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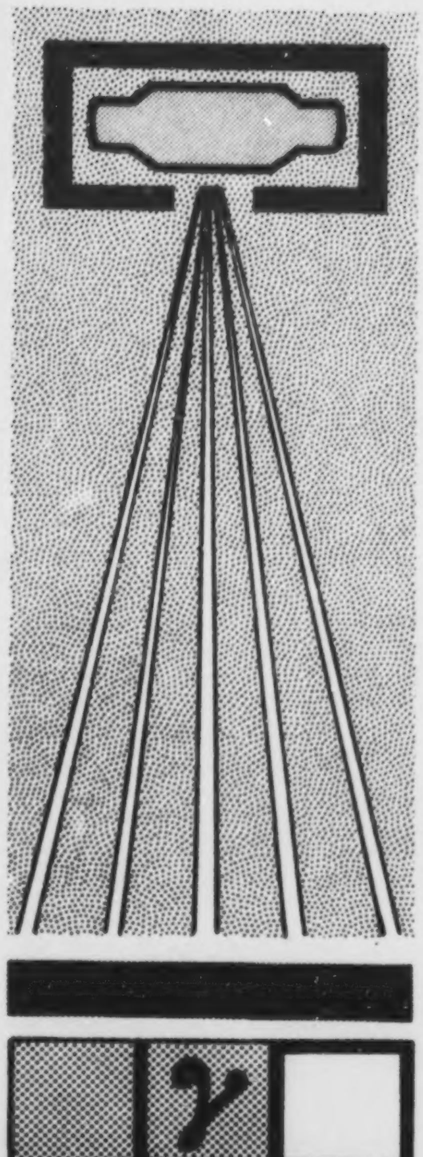
A COLLECTION OF RADIUM LEAK TEST ARTICLES



A COLLECTION OF RADIUM LEAK TEST ARTICLES

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
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MEDICAL AND OCCUPATIONAL RADIATION PROGRAM

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MORP 68-1

A COLLECTION OF RADIUM LEAK TEST ARTICLES

Assembled by
VALORIE A. WOOD
Radioactive Materials Section
Medical and Occupational Radiation Program

MARCH 1968

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service

Bureau of Disease Prevention and Environmental Control
National Center for Radiological Health
Rockville, Maryland 20852

PREFACE

Contained in this collection are known articles related to methods of radium leak testing, which have appeared in the open literature or are in preparation for publication. Established methods of leak testing, as well as new and untried methods, are included. The methods described are designed for use both in the field and in permanent installations.

It is hoped that the compilation will serve as a convenient reference for persons interested in information on radium leak testing.

Permission to reproduce the articles has been received from the authors and the journals.

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Apparatus and Method for Testing Radium or Thorium Capsule Seals

R. J. Beal
Health Radiation Branch, National Research Council, Atomic Energy Project, Chalk River, Ontario

February 6, 1950

SEALED radium capsules, after equilibrium has been reached, contain radon and radon decay products. Similarly, thorium capsules contain thoron and thoron decay products when equilibrium conditions have been established. In both cases the decay products or active deposits are solids which attach themselves to dust particles. The active deposits are strongly alpha-active, the radium active deposit having a half-life of about 50 minutes and the thorium deposit a half-life of 10.5 hours.

The method here described depends upon the fact that the source capsule while under test, will expel a part of the emanation and accompanying decay products, if the seal is faulty. The expulsion of the emanation is achieved by heating the capsule by a steam jacket to about 100°C. It has proved convenient to collect the active deposit by carrying out the heating with the source placed in the collecting volume of an electrostatic precipitator. The particulate activity precipitates on the inner surface of a removable cylinder, which forms the outer electrode of this precipitator assembly. The activity could no doubt be collected by the long established method of collection on an electrode charged negatively to a few hundred volts. Counting has been done by making the collecting cylinder the outer electrode of a methane-filled cylindrical proportional counter. A very high counting efficiency is thus secured.

The apparatus, as shown in Fig. 1, consists of a cylindrical precipitator with a central electrode at 10 kv positive potential relative to the collecting cylinder. The source under investigation is placed in a cup which slides into position at the base of the cylinder. The complete precipitator and source assembly is surrounded and supported in a steam jacket. The steam jacket is carefully sealed from the precipitator assembly to ensure that moisture does not interfere with precipitation.

The first step in the procedure consists of placing the source in the source cup which is then elevated and locked in position in the precipitator base. Then the steam is turned on and allowed to circulate during the entire time of the test. High tension is applied to the h.t. electrode for 90 minutes. The equipment described above requires about 30 minutes to reach operating temperature of 90°–100°C and a total testing time of 90 minutes is used so that an hour of precipitation occurs at maximum temperature of the test. The collector as used fits into a methane proportional chamber for counting.

Some typical results secured with the method outlined above are given.

(*a*) *A radium-beryllium source of 200-mc nominal value.*—This source gave a precipitation which counted 1500 alpha-c.p.m. It is interesting to note that when this source was held in the apparatus at 10 kv for 24 hours *at room temperature*, only 30 c.p.m. were collected on the tube.

(*b*) *A radium needle of 1-mc nominal value.*—This source gave a precipitation which counted 77 alpha-c.p.m.

(*c*) *Sources ranging in value from 1 mg to 1.2 g.*—A number of sources, ranging in value from 1 mg to 1.2 g, were checked and most of these, when subjected to the test, gave counts within 10 c.p.m. of the normal background of the counter, indicating a satisfactory source seal.

(*d*) *Effect of radium contamination on the exterior of the capsule.*—Monel metal cylinders approximately the same size as a source capsule were prepared with surfaces varying from a high polish to an extremely rough, pitted surface. These monel cylinders were heavily contaminated with radium sulfate. On precipitation only the cylinder with the roughest surface gave any appreciable counts above the normal background, namely 5 c.p.m. Thus, it would appear that surface contamination under normal capsule conditions has little or no appreciable effect on the results of the test.

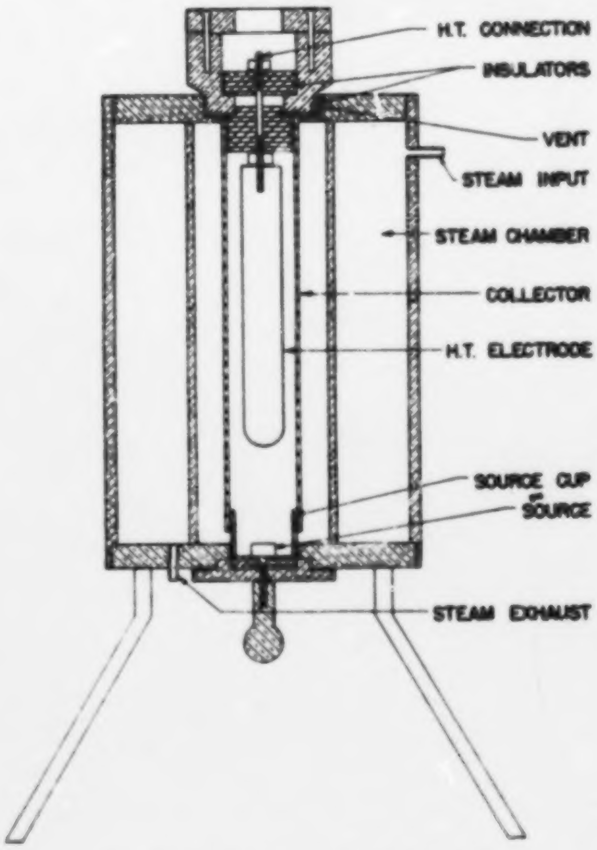

FIG. 1. Radium and thorium capsule seal tester.

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PHOTOGRAPHIC TECHNIQUE FOR LEAK TESTING RADIUM SOURCES*

By PAUL H. BENROSIAN, Ph.D
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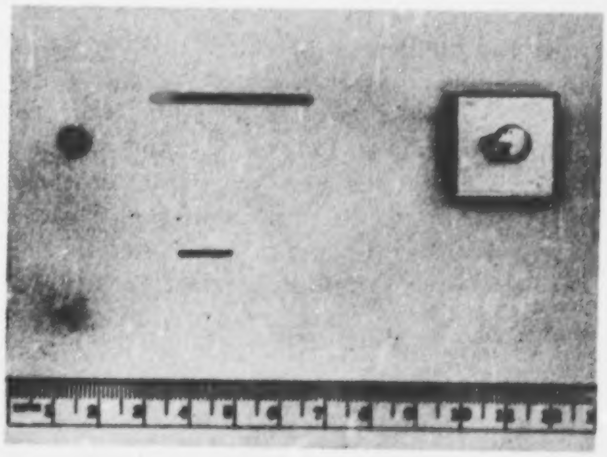

Fig. 1. Examples of sealed sources of radium 226 as shown from left to right include: 2.5 mg. plaque; 15 mg. needle; 5 mg. cell below; and 10 mg. plaque

TO USERS of sealed radium 226 sources and to personnel of radiation control agencies, who are responsible for public health, a quick, reliable method for determining if sources are leaking is extremely important. As the frequency of testing the sources (Fig. 1) for leakage is increased, the probability of discovering faulty ones and being able to take proper action before radioactive contamination or health hazards occur is also increased. Unfortunately, no rapid, dependable method exists for leak testing radium 226 sources. Many of the current techniques generally require that the source be placed in a leak tight chamber for periods up to 24 hours, after which time the chamber is examined for radium and daughter products resulting from the leak. Examples of such tests include the Hale[3] method in which the leaking source is placed in a stoppered test tube containing 5 ml. of tap water with 0.37 gm. of activated charcoal. After 24 hours, the source is removed and 24 hours later the stoppered test tube is placed in a well scintillation counter.

Other techniques which offer variations of the Hale method include the cotton plug and jar test systems.[1,4] In the first of these 2 procedures, the leaking source is placed in a sealed test tube or bottle for 24 hours with a wad of cotton. After this period, the cotton is removed and measured for radioactivity by a G-M survey meter. In the jar test, the inside surface of the screw-top lid is examined with an alpha survey meter after the source has been sealed in the jar for 24 hours.

Gallaghar and others perfected an electrostatic technique.[2] In this method the radium source is placed in a sealed cylindrical chamber having electrodes at either end across which a 300 volt DC potential is applied. Because radioactive decay of radium tends to create positively charged daughter products, most of these products are deposited on the cathode where they can be measured by a G-M counter. The period of collection ranges from a few minutes to several hours.

These techniques are hindered not only by the long period required for sample collection, but also by the need for either radiation survey meters or laboratory detection instruments to determine the presence of radioactive leakage. In addition, results from many of these tests are frequently unreliable and may be misleading.

Because of the disadvantages in current leak testing methods, the Radium Technology Unit at the Southeastern Radiological Health Laboratory has conducted preliminary investigations of a photographic method. The essence of this new procedure is the ability of alpha particles to interact with a zinc sulfide (ZnS) scintillator, thereby producing light which can be recorded by high speed photographic film.*

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* From the U. S. Department of Health, Education and Welfare, Public Health Service, Southeastern Radiological Health Laboratory, Montgomery, Alabama.

In the decay chain of radium several alpha, beta and gamma emitters are produced. Since alpha particles cannot penetrate the walls of a sealed source, it is obvious that their detection external to the source is evidence of a leaking or contaminated source. One way, therefore, of determining if a leak or contamination exists, is to look for alpha emitters outside of the source. This can be done almost instantly with the photographic technique.

EXPERIMENTAL

As the major objective of the photographic technique is to detect light produced only by the alpha-ZnS interaction in a mixed radiation field, it was necessary to evaluate the extent to which beta and gamma radiation might also produce light. For this purpose, experiments were conducted using 2 leaking Ra^{226} plaques, 2.5 mg. and 5.0 mg., and a 3 mc Co^{60} source. Each source was tested separately by placing the source in a 2 inch glass Petri dish, which is opaque to alpha particles, and placing the dish on a piece of ZnS coated plastic screen with the ZnS facing the source. The source, dish, and ZnS were arranged with a simple lens and a Polaroid 4×5 Land film holder assembly so that the image formed on the ZnS sheet by the radiations from the source could be photographed. The experiments were repeated with the sources placed directly on the ZnS without the Petri dish. Comparison of the images showed conclusively that the ZnS emitted light not only when it was exposed to alpha particles but also to beta-gamma radiations and particularly to beta. The comparison, however, also indicated that the alpha contribution in an alpha-beta gamma field could be easily discriminated by a shielding material. In addition, experiments were conducted to improve the scintillator's response to alpha particles in the presence of beta radiation. The ZnS screen, which was obtained commercially and which contained about 10 mg. ZnS salt mixture/cm.^{2}, was treated with 12N HCl for 30 minutes or until the salt concentration on the screen reached 5 mg./cm.^{2} This modification of the scintillator reduced the beta effect on the ZnS, thereby highlighting the alpha contribution.

Based on these preliminary findings, a more efficient photographic system was devised which included a device to properly orient the source to the ZnS and film. A schematic diagram of this system (Fig. 2) is shown together with explanatory terms.

In operation, two foot tongs are used to place the source in the 20 pound lead sample chamber. The source is laid on the alpha absorber (plastic-8 mg./cm.^{2}) overlying a ZnS screen which rests on a thick piece of plexiglass (1.6 gm./cm.^{2}). This

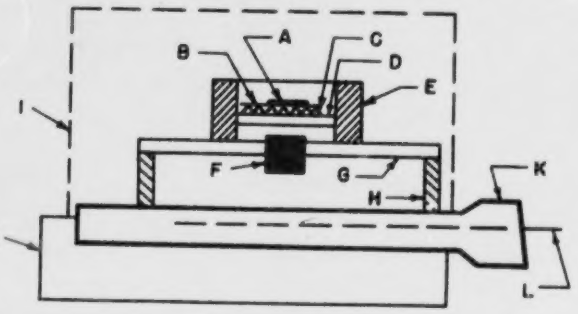

FIG. 2. Schematic diagram of the photographic lens system showing the lead sample chamber in relation to the lens and the film holder.

The various components of the system are as follows:

A. Source
B. Movable alpha absorber (plastic—8 mg./cm.^{2})
C. Movable ZnS screen
D. Plexiglass sample support (1.6 gm./cm.^{2})
E. 20 pound lead shield sample chamber assembly
F. 10× ocular lens
G. Sliding wooden platform
H. Supporting tracks for platform and chamber
I. Light-tight lid
J. Wooden base
K. Film holder
L. Film—4×5 packet

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* For the experimental studies in this investigation, Polaroid Land film ASA 3,000 type 57 in the Polaroid 4×5 Land film packet system was selected. The film holder in this system is lightweight and its cost is under 50 dollars. Use of this equipment does not constitute an endorsement by the Public Health Service.

chamber assembly is supported by a sliding platform and rests directly over a 10× microscope ocular lens. The chamber, platform and lens are fixed together and may be moved from side to side as a single unit over the film holder. When the source is in place, the light-tight lid is closed and a 4×5 film packet is slipped into the film holder. In making the exposure, the protective envelope of the film is pulled outward from the holder but not removed completely from it. The plastic shield over the ZnS prevents the alpha particles from interacting with the ZnS and the film records light produced only by the interaction of beta and gamma radiations with the scintillator. The light is focused through the lens and a well defined image is formed on the film. The protective envelope is replaced, the light-tight lid opened and the sample platform moved over to the next exposure position. In this new position, the plastic shield is removed and the source is placed in direct contact with the ZnS surface. The exposure procedure is repeated for the second picture. In this manner, 2 different exposures are made on the same film. (By slight alterations in the design of the sample platform, many more exposures can be made on a single film.) For the shielded situation, only the beta-gamma interactions on ZnS are produced. In the unshielded case, the alpha and beta-gamma effects are photographed. By comparing the 2 images, the alpha contribution can be seen. This technique, which requires about 3 minutes for each exposure and 10 seconds for development of the print, can detect point sources of radium contamination or leaking radium or exposed radium as low as 500 picocuries in medical sources ranging up to 50 mg. of radium. The limit of detectability was evaluated experimentally by photographing various levels of unsealed radium in conjunction with different sources. In addition, dosimetric measurements indicated that a person using the photographic technique in testing ten 10 mg. needles during a period of 1 hour would receive about 2 mr whole body exposure and about 15 mr to his hands.

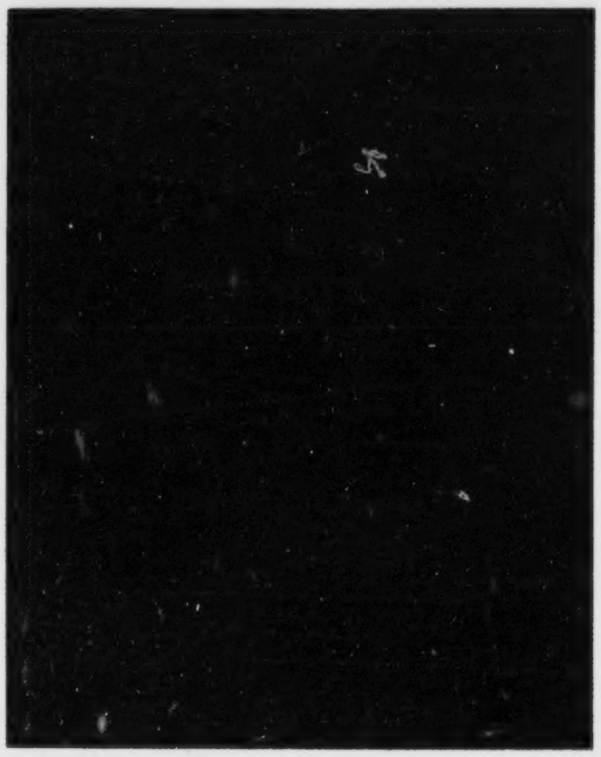

Fig. 3. A dual exposure of a 5 mg. cell which registered essentially no alpha activity in the jar test determination. The exposure time was approximately 3 minutes for each picture. For image on left, the cell was unshielded. For image on right, the cell was shielded from the ZnS by an alpha absorber (plastic—8 mg./cm.^{2}). The images are practically alike and, therefore, indicate no radium leakage or contamination above 500 picocuries, the detectable limit of the photographic method.

Interferences in the photographic method were found to be minor. No significant amount of direct film exposure resulted from the gamma rays associated with the leakage material or the source itself. This was probably due largely to the position of the source during an exposure (about 11 cm. above the film) and of the film's relative insensitivity to gamma radiation as corroborated by Co^{60} irradiation experiments. Beta radiation from the source, however, did cause general fogging of the film. This effect was eliminated by using a thick piece of plexiglass (1.6 gm./cm.^{2}) as the sample support.

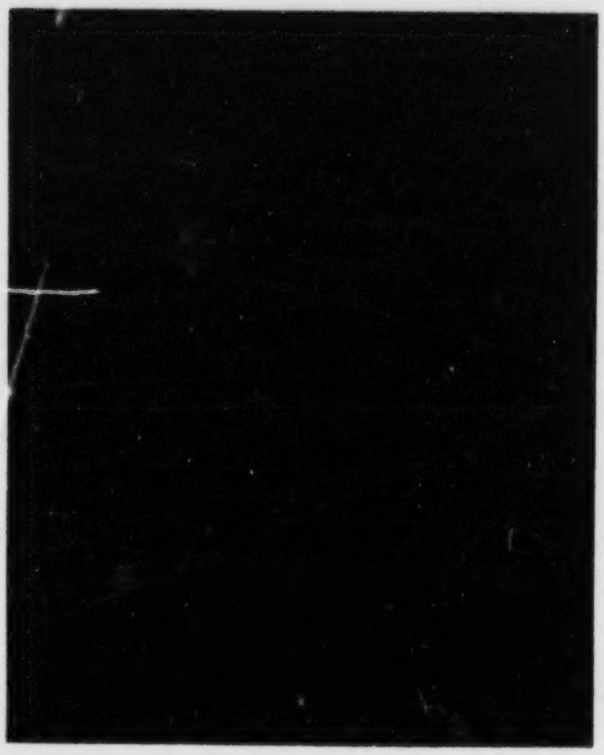

FIG. 4. Exposures of an unshielded and shielded leaking 2.5 mg. radium 226 plaque which showed an alpha activity of 500,000 cpm by the jar test. The 3 minute exposures show a marked difference in the images as well as the location of point sources of exposed radium and daughter products on the plaque.

RESULTS AND CONCLUSIONS

Qualitative experimental results can be readily seen in pictures beginning with Figure 3. The image on the left in all the leak-test photographs represents the effects of all external radiation produced by the source and the right hand image is formed by all external radiation except alpha. The significant difference between the 2 images, which are magnified approximately 1 1/2 times, is due, therefore, to the alpha radiation as produced by a leaking or contaminated source. As it applies to the photographic method, a leaking source is one which is releasing or exposing radium 226.

As an example of what a nonleaking radium source (Fig. 3) would look like, a 5 mg. cell was used in the unshielded and shielded positions. It is evident in comparing these pictures, that there is little or no difference between the images. From this observation, it is possible to conclude that the source is not contaminated or leaking radium, at least above the 500 picocurie limit as established for the photographic method. In contrast to this, a 3 minute exposure (Fig. 4) of a 2.5 mg. radium plaque, which registered over 500,000 cpm by the jar test, depicts the leaking and contaminated nature of the source quite well. The reverse side of the 2.5 mg. plaque (Fig. 5) which was purposely contaminated shows the distribution of contaminants despite the strong beta-gamma background.

A badly leaking 5 mg. plaque was also tested (Fig. 6) and the results are quite conclusive. This particular source was an obvious leaker because it was noticeably damaged. There are, however, many sources which look good and structurally sound, but leak just as badly as visibly

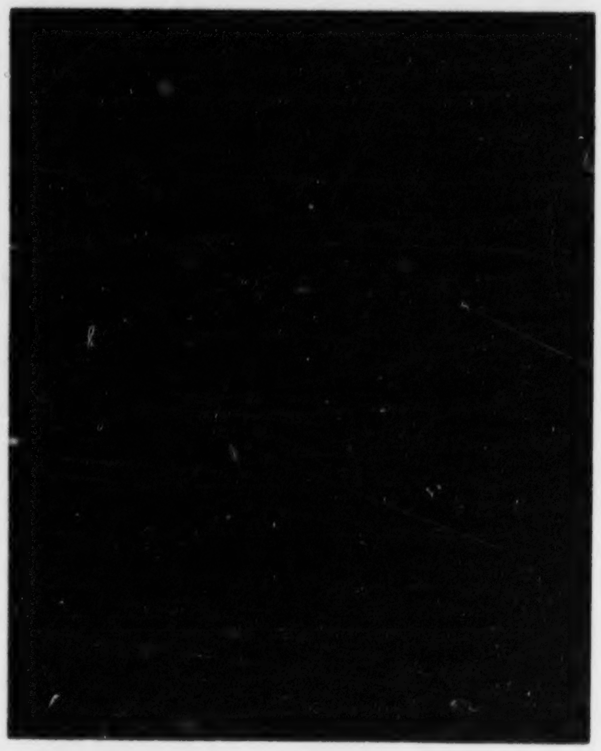

FIG. 5. The contaminated reverse side of the plaque shown in Figure 4. The points of contamination are easily seen in comparing the 3 minute exposures.

broken ones. Two such apparently good cells were examined (Fig. 7 and 8) and found to be leaking or contaminated at various positions along their lengths. Both cells contained about 3 mg. of radium and one which appears badly damaged (Fig. 7) showed about 4,000 cpm in the 24 hour jar test, whereas the other indicated only 300 cpm by the same test.

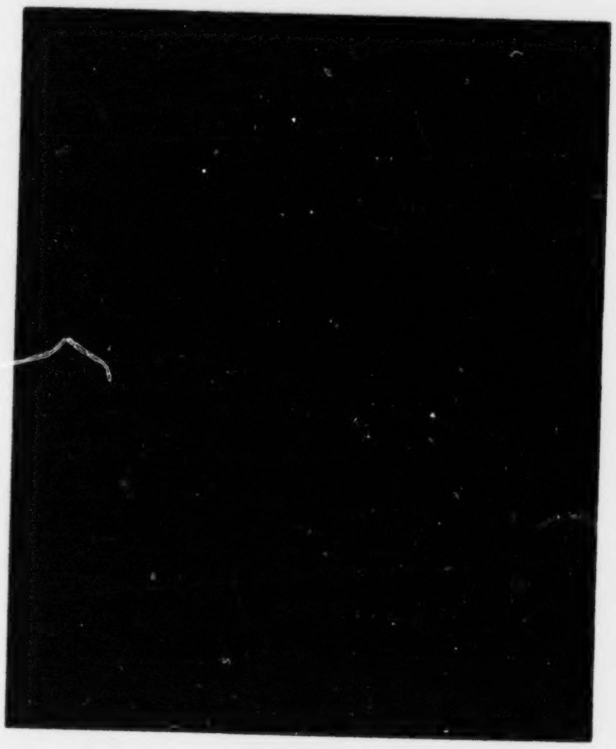

Fig. 6. An extreme difference between the two images is apparent in these 3 minute exposures of a badly damaged and leaking 5 mg. plaque.

Fig. 7. These 3 minute exposures were made of a 3 mg. cell which appeared to be in perfect condition. The significant difference in the images clearly indicates the leaking or contaminated nature of the source and the area of damage. When measured by the jar test, this source produced 4,000 cpm of alpha activity.

Studies are continuing at the Southeastern Radiological Health Laboratory to improve the picture quality of the photographic technique, to improve its sensitivity, and to automate the device so that it may be operated with less handling. Additional experiments are in progress in the development of a system which can more easily distinguish leaking sources from those that are only contaminated. The photographic method in its present state represents a qualitative leak testing means which can be applied to several facets of radium usage. It has been used to distinguish sources having various amounts of radium from each other and also for detecting radioactive contamination picked up in wipe tests. With proper backlighting (Fig. 9) the photographic system can be easily adapted to assess the distribution of radium within a source. Physicians, radiological health personnel and others will be able to check the integrity of radium sources more frequently and rapidly than is currently possible. Physicians, for example, can quickly examine their sources before and after use in patients and determine at least if they have dangerously leaking or ruptured sources. Since no special instruments are needed to make leak tests by this method, sources can be examined at little cost and readily evaluated at the hospital or in the physician's office. It is also possible to test several sources or make several exposures on a

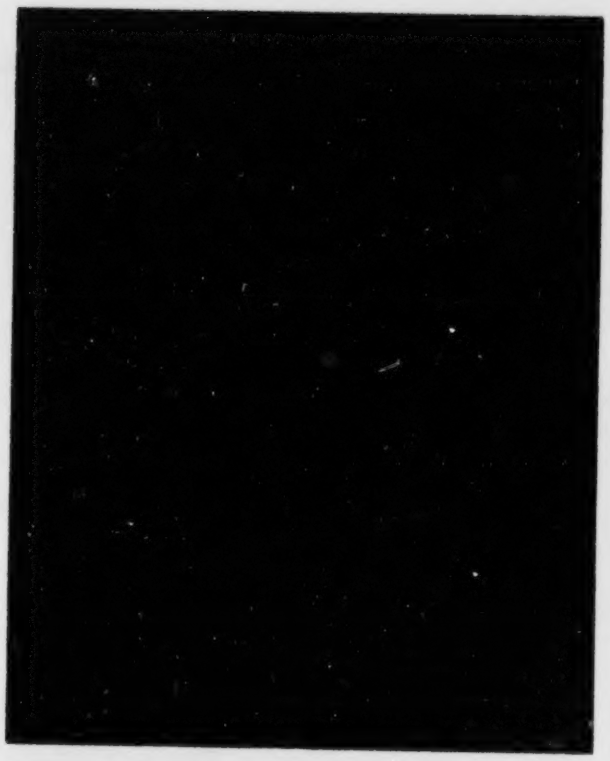

FIG. 8. This 3 mg. cell is similar to the one used in Figure 7 but, as these exposures indicate, the leak is different and is confined primarily at one end. This source produced only 300 cpm of alpha activity when examined by the jar test.

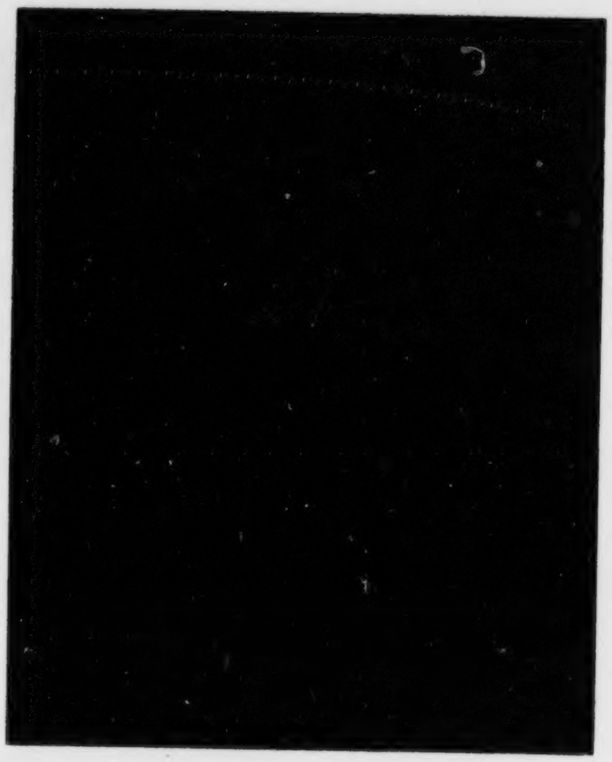

FIG. 9. With proper backlighting, the photographic technique can be easily adapted for use in locating radium 226 within a source. The above picture shows the distribution of radium within a 5 mg. cell.

single film. Contamination of the photographic components or of the sample chamber by leaking sources can be avoided by enclosing the film holder in a disposable plastic bag and by lining the chamber with disposable plastic sheets.

SUMMARY

The photographic method provides a simple, positive and economical means by which radium sources may be regularly and frequently tested for radium leakage and contamination by the user. In this manner, badly leaking sources may be discovered before they cause widespread contamination of buildings, storage facilities or other areas where radium 226 is stored or used. Because of its versatility, its rapid operation and ease of use, the photographic technique represents a safe, effective system which can be used by medical and radiation control personnel for protecting physicians, patients and others against unnecessary radiation exposure.

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9

A TECHNIQUE IN TESTING RADIUM SOURCES FOR LEAKAGE*

By JAMES S. BENSON *and* RICHARD H. FETZ†

SEALED sources of radioactive material, including those that are used in medicine, are subject to possible leakage, which may create a significant health hazard. To identify leaking sources, the International Commission on Radiological Protection (ICRP) recommends periodic leak testing of all sealed sources of radioactive materials, including radium.[5] Once found, a faulty source should be disposed of or re-encapsulated.

Radium is encapsulated as a powdered salt which if released could be easily spread over wide areas. For example, one ruptured source could contaminate an entire hospital, necessitating decontamination because of the high radiotoxicity of radium.[2] Clean-up operations for such an incident would be difficult, and the resulting costs could be prohibitive. Through periodic leak testing of radium sources and the disposal of leaking sources, the probability of widespread contamination can be reduced.

Leakage of radium salts and/or radon 222, the chemically inert gaseous daughter of radium, may result from damaged or defective sources. Although radon is less hazardous biologically than radium, radon leakage is an indication of poor source integrity and may be a warning signal for the more serious leakage of the radium salts. Therefore, a radium leak test method should be sensitive enough to detect radon leakage.

In an effort to find a suitable means for leak testing sources on location during a statewide survey of radium users in Georgia, the authors selected from the literature the electrostatic, charcoal-water, and cotton plug methods for experimentation.[3,4,6] The electrostatic and charcoal-water methods required too much time for loading the sources into and removing them from the leak test apparatus. This source handling problem would tend to increase the exposure to the person performing the leak test. The problem would be accentuated when many facilities with large radium inventories were surveyed, and all the radium at each facility was leak tested. Although the cotton-plug method required less handling time, problems of reproducibility were encountered. It was obvious that a method that would allow rapid source handling, while not sacrificing reproducibility and sensitivity, was needed.

METHOD

A method that met these criteria was developed. It is based on the assumption that when a leaking radium source is placed in a sealed container, the gaseous radon leaking from the source will diffuse throughout the volume of the container, and the particulate daughter products of radon will deposit on all surfaces (an observation first made by Madame Curie[1]). Then, a measurement of the activity on the inner surface of the container lid, an area of standard size that allows a constant counting geometry, will give a quantitative reproducible measurement of radon leakage.

The length of time the sources are in the container affects the amount of activity collected because of radon growth. For example, if leakage is constant, a collection time of 1 hour permits the radon to build up to 0.75 per cent of its equilibrium activity, 4 hours to 3.0 per cent, 8 hours to 5.9 per cent, 24 hours to 17 per cent, 48 hours to 30 per cent, and 30 days to 100 per cent. Field tests show that a 24 hour collection time will provide sufficient sensitivity with minimum inconvenience.

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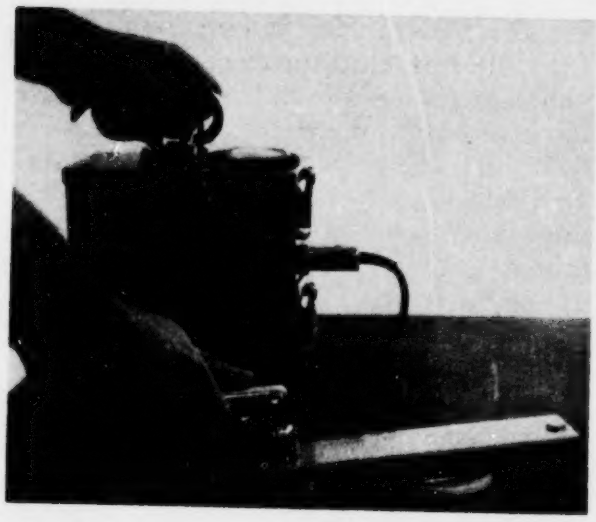

Fig. 1. Checking for alpha activity on jar lid.

After the 24 hour collection time, the container lid is removed, and the probe of an alpha detection instrument is immediately placed over its inside surface (Fig. 1). Although other instruments may be used, alpha instruments offer greater alpha sensitivity and are less sensitive to surrounding gamma fields. Radon daughter activity on the lid indicates radon leakage from either inside the source or from radium salt contamination on the source. It is important that the reading be taken immediately after removal of the lid because the radon daughter activity decays with approximately a 30 minute half life.

Various types of containers such as Petri dishes and metal ointment cans were evaluated as leak test containers. An ordinary glass jar with a rubber gasket seal on the lid to prevent the escape of radon from the container produced the best results. The inner dimensions of the jar used were 2⅛ inches in diameter, 1⅛ inches in height, and ⅜ inch recess on the lid. These dimensions made it possible to place sources of various sizes and shapes in the jar.

As a theoretic illustration of the sensitivity of this system, assume that 0.001 μc of unsealed radium 226 is placed in a jar as described. After 24 hours, approximately 175 disintegrations per minute alpha activity from radon daughter products would be present on the jar lid under the following conditions: (a) 100 per cent of the radon were released by the source, and (b) 100 per cent of the radon daughter products were uniformly deposited on the inner surfaces of the jar.

The use of beaker tongs to handle the jars and 6 inch forceps to handle the radium sources enables the tester to perform the leak test with minimum exposure to the hands (Fig. 2). The use of lead bricks, lead blocks, and lead safes for shielding will reduce gamma exposure to personnel during leak test periods.

CONCLUSION

The jar method was used to leak test radium sources in 21 hospitals in Georgia. A total of 3,388 milligrams of radium contained in 725 sources was leak tested over a 9 month period. Of the 21 hospitals visited, 10 had sources indicating leakage. No exposure higher than 30 mr per month

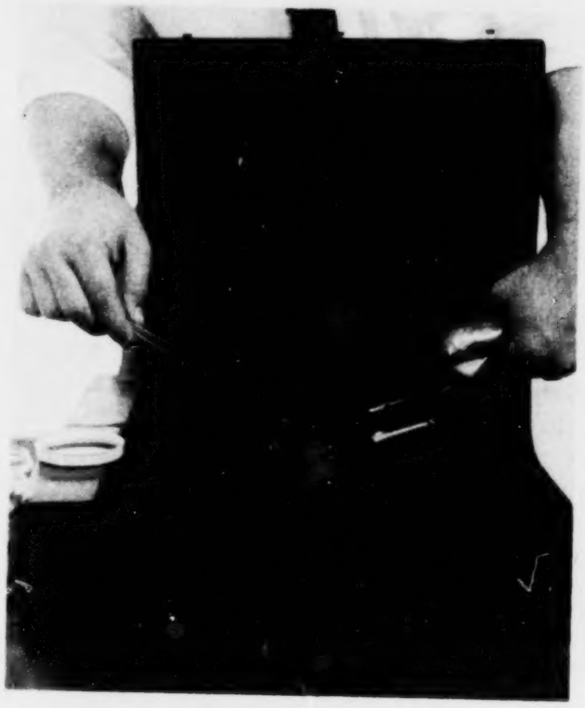

Fig. 2. Placing radium sources in jar.

was indicated by the wrist and waist film badges worn by surveyors during the testing period.

The Georgia Department of Health is continuing the use of the jar method, and it is now being used by personnel in several other State health departments.

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13

A TECHNIQUE FOR AUTO-RADIOGRAPHS OF RADIUM CONTAINERS

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ATTENTION was drawn by Russ and Clephan in the *Lancet*, August 9, 1930, to inequalities in the distribution of radium in tubes and needles as shown by auto-photographs or auto-radiographs, which seems a more exact term. At intervals of five to ten years all radium containers ought to be tested in this simple way to make sure that the distribution remains as it is intended to be: it is obvious that if, for instance, all the active substance in a series of tubes should be evenly distributed and is not, inaccuracies will result in calculated and carefully planned treatment. Modern treatment sometimes demands a greater concentration of the radioactive substance at the two ends of the tubes for providing more homogeneous irradiation, and a check on the distribution is needed from time to time. It seems worth while therefore to publish an account of the technique devised in the Barnato Joel Laboratories, with elementary details which may be of use at hospitals having no physicist but facilities that would enable any radium containers in the possession of such hospitals to be auto-radiographed on the premises. Anyone with a good working knowledge of photography can produce good pictures if they carry out a suitable technique, but for their own sake, it is essential that they should have a thorough understanding of the dangers of handling radium.

Although the principle of auto-radiography is perfectly well known to people who work with radium and X rays, it should perhaps be stated briefly for the sake of those who do not. Auto-radiography is based on the fact that the rays from radium, like X rays, fog photographic plates and films as though they had been exposed to light. These auto-radiographs are produced by placing radium containers in contact with plates or films for such time as will allow the radium to fog them enough to show a picture of the distribution of the contents when they are developed. Containers may be in the form of applicators, tubes or so-called needles, but both tubes and needles are in tubular form and both usually have eyes for threading.

To see the distribution of the radium inside the tubes we must have a picture of the tubes as well as the contents, and this can be done either by giving the X-ray film a minute dose of X rays or by a flash of light during the time that the tubes are in contact with the film, the tubes being between the film and the source of light or X rays. The film is thereby fogged except for those parts of it that have lain in contact with the tubes and have thus been screened from the light. The net result should show light tubes upon a slightly fogged background, the contents of the tubes showing up clearly and darkly inside them; but when the film is printed the tones are of course reversed.

We have not used the method of laying the tubes upon wrapped X-ray films in daylight under a small X-ray bulb and giving a minute flash of X rays while the tubes are *in situ*. With our technique the radium tubes are laid upon unwrapped films in the dark room, the flash being provided by an automatic electric contrivance devised by S. B. Osborn. This is a convenient piece of apparatus and the small light, which is hung exactly above the film at a distance of one and a half metres, goes out automatically a second after the button has been pressed. On the lighting theme many variations are possible: people dealing with only a few tubes could use an electric torch held above the film, but it would be necessary to experiment a little so

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as to get a suitable tint for the background and there would be no certainty of uniform results. The fogging is easily overdone and this makes a bad picture. It does not matter where the source of light is so long as the radium is between it and the film: it might just as well be at the bottom of a cone or enclosed rectangular space with a top of clear glass on which the radium could be placed with the film on the top of it for exposure. In fact if it were necessary to auto-radiograph many grammes of radium it would be well worth while to have some such apparatus specially designed and made with an adequate thickness of lead incorporated, extending from the ground and above the level of the shelf or table top so as to give protection to the body of the person doing the work.

Apparatus required

1. The best lead protection available for the persons who are identifying and auto-photographing the tubes.
2. Long forceps. It is wise for one's own sake to get accustomed to handling radium with long forceps, although they are not so handy as smaller ones.
3. A powerful lens to make identification of the tubes as easy and swift as possible.
4. Wooden blocks with lines of holes drilled in them of a suitable depth and bore to hold the tubes in numerical order after identification. The designation of the series should be marked in pencil on the block and its number below each tube. These marks are easily erased.
5. A series of small opaque numbers for the films.
6. Wratten safelight 6A or 6B.
7. Electric torch or low-powered bulb.
8. X-ray films.
9. Standard X-ray developer.
10. Ordinary darkroom facilities.

It is essential to have the following specifications of the tubes for on these the exposure is calculated—content, active length, screenage (or wall thickness), and diameter of bore. In these laboratories the tubes are quickly examined and identified from behind a thick lead-lined L-shaped screen in the radium strong room, with a good light and powerful lens, and arranged in sequence in the holes of the wood blocks provided for the purpose. The blocks are carried to the darkroom in lead-lined boxes. We make it a rule not to have more than a few hundred milligrammes in the darkroom at one time, partly to minimise personal exposure and partly on account of the photographic plates and papers stored there which would in time be affected by the rays. Recently when tubes containing a total of over four grammes were auto-radiographed, all plates and papers were removed to another room, but this is unnecessary when dealing with comparatively small quantities.

The three chief difficulties in working out the technique are the choice of film, the estimation of exposure, and the fact that the tubes are inclined to roll when they are placed upon the photographic film.

When long series of tubes have to be auto-radiographed one must have some means of fixing them in numerical order on something that can be lifted and laid upon the film for the length of time needed to give a suitable exposure. There is a double reason for this—the job can be done more quickly, speed being one of the first essentials, and such a gadget prevents the tubes from rolling during exposure. If, however, the amount of radium to be dealt with is small it is perhaps hardly necessary to spend time in making apparatus of this kind. The simplest method is to lay the tubes upon the photographic film, then give a flash of light and remove them when the exposure of the radium is complete. The number auto-radiographed at one time is necessarily limited by the length of exposure required for the particular type of tube: for instance, with those of very high intensity needing a few seconds only, only one could comfortably be exposed at a time; whereas if they are of low concentration with about 60 seconds optimum exposure, at least half a dozen could be laid upon the film with 10 seconds interval between each. Ten seconds would then be left in which to give the flash, after which the exposure of the first would be complete and they could all be removed after the same intervals. If the exposure is only for a few seconds, a tube that rolls before it comes to rest upon the film will produce a blurred effect and the exposure will have to be repeated.

Exposure trays

For exposing large quantities of radium we use trays consisting of clear film, in which slits have been cut to hold the tubes, stuck to brass frames. These are of brass rather than wood to avoid warping, and the sheets of clear film must remain absolutely flat if the tubes upon them are to be in contact with the photographic film and produce a clear picture. The frames are made slightly larger than the photographic films we use so as to be able to see easily by the safelight the position of these through the trays when they are loaded and put into position for exposure. Some patience and dexterity are needed in cutting the slits; it is easy to see that the tubes must fit snugly so that they do not roll or drop through when the trays are lifted. Cutting is most easily done over glass with a sharp razor blade or scalpel and a small steel straight edge. It is a help in exactitude to mark out on millimetre squared paper the required holes just large enough to hold the tubes: this paper should be placed between the clear film (ordinary X-ray film, cleaned, will do) and the glass, and all should be firmly strapped together at the corners for the cutting process. We found a thin coating of seccotine the best adhesive: the film is apt to buckle, particularly if the slits are long and numerous, and it is well to keep the tray in a press until it is well stuck. It matters little if shorter tubes are placed in longer slits provided that the width is correct. (Fig. 1.)

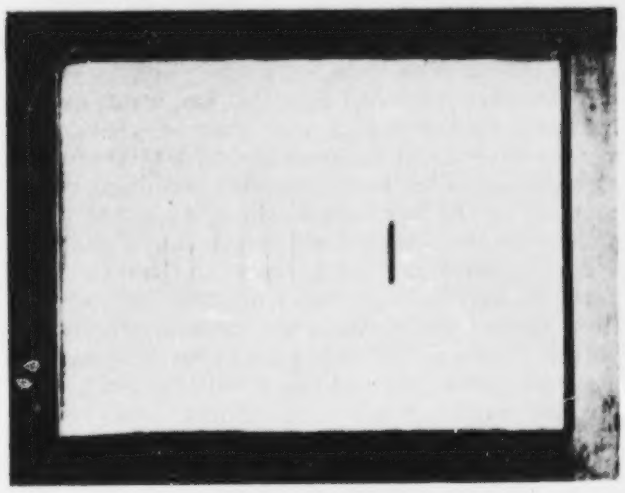

FIG. 1.
Exposure tray with radium tube lying in one of the slits.

The trays can be loaded and the numbers placed upon them in the darkroom with the protection of the lead L-piece before the ordinary light is turned off. X-ray films are duplitised, that is, both sides are covered with emulsion, but the opaque numbers, placed upon them and imprinted by the flash of light, serve to show which is the contact side when they are developed.

The choice of films

As speed is important it is best to use duplitised X-ray films which are, of course, more sensitive to the rays than ordinary photographic plates or films which give excellent clear pictures of radium tubes but require much longer exposures. The Ilford Company produce two grades of films, "Ilford" and "Ilfex", and we have used both: Ilford need about double the exposure given for Ilfex to get the same result, but are more easily fogged with faulty technique. Kodak films would no doubt be equally good. The constant in the formula worked out for exposing tubes of varying quantity, active length, diameter, and filtration could be altered to suit the films chosen. It is important to ~~use a suitable~~ *safelight* (either 6A or 6B) when exposing and developing these films and to turn the light away when doing any part of the proceedings such as development for which no light is needed.

Using the formula given on page 120 to calculate exposure, good results should be obtained for Ilford or Ilfex films with a 4 to 5 minute development at about 65°F.

Exposure

This was done at first by trial and error: the factors involved in the different categories of tubes were roughly estimated and experience soon produced good pictures. When large numbers of tubes of different content, active length, active diameter and filtration had been auto-radiographed the resulting data were considered by C. F. Walker and W. A. Jennings, to whom I am indebted for the following exposure formula for Ilfex films, development 5 minutes at 65°F.

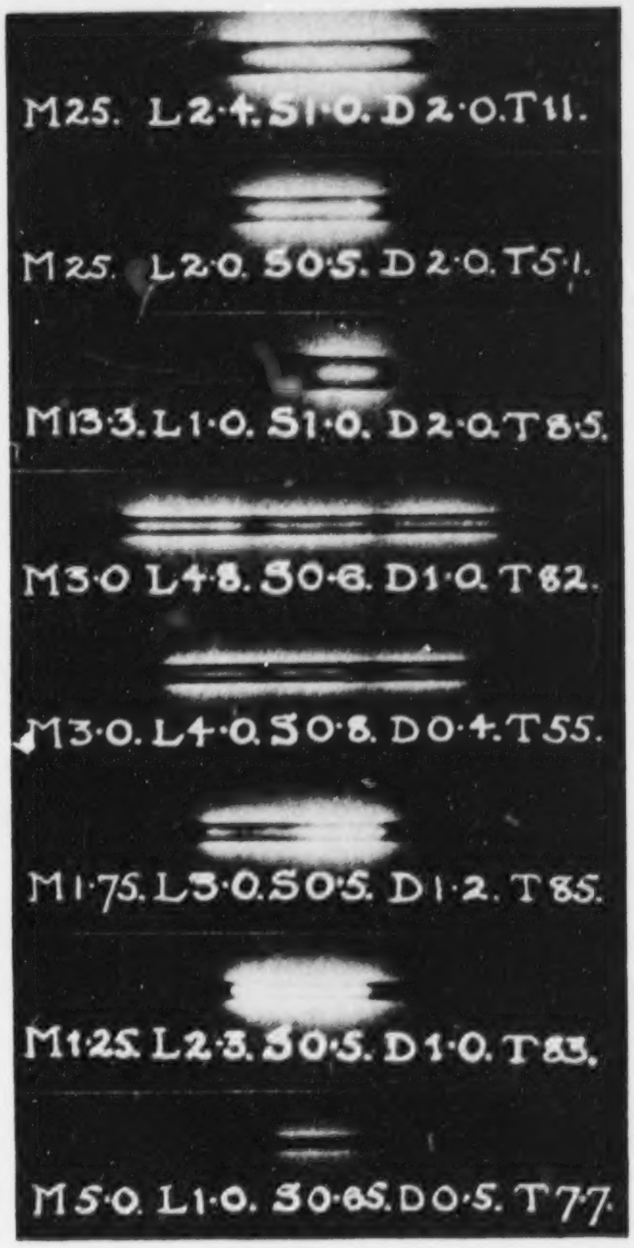
M25. L2·4. S1·0. D2·0. T11.
M25. L2·0. S0·5. D2·0. T5·1.
M13·3. L1·0. S1·0. D2·0. T8·5.
M3·0 L4·8. S0·6. D1·0. T82.
M3·0. L4·0. S0·8. D0·4. T55.
M1·75. L3·0. S0·5. D1·2. T85.
M1·25. L2·3. S0·5. D1·0. T83.
M5·0. L1·0. S0·65. D0·5. T7·7.

FIG. 2.

Records of all the following specifications should be available—without them exposure is a matter of guesswork.

Radium content in milligrammes .. = M
Active length in centimetres .. .. = L
Screenage or wall thickness in millimetres .. .. = S
Diameter of bore in millimetres .. = D
The calculated time of exposure in seconds .. .. = T

$$T = \frac{10\,(1 + S)^2 \times L\sqrt{D}}{M}$$

For the slower grade Ilford films, the constant 10 must be doubled. The formula serves as a working basis for exposing any radium tubes or so-called needles in clinical use, the constant being adjusted to suit the speed of the film used. It was found that if a full development was given with Ilfex films, the exposure for certain high intensity tubes was so short that it was difficult to manage. The accompanying auto-radiographs (Fig. 2) were therefore made on Ilford films with a constant of 20 for variations in M (1·25–25 mg.), in L (1–6 cm.), in S (0·5–2·0 mm. platinum) and D (0·4–2·0 mm.)

In Fig. 2 for the purpose of illustration the various specifications and the time of exposure worked out from them were written on the film, instead of the numbers engraved on the tubes, before it was printed.

The radium should be handled in the darkroom, as in the strong room, from behind a lead L-piece and the film should also be placed behind this for exposure, with the light for the flash (if a fixed bulb is used) hung exactly above it.

Identification of the tubes upon the films

Some care is needed over this, but it can be done correctly by keeping an exact record of the tubes as they are laid out for exposure, with their numbers appended. The small opaque numbers corresponding to the numbers in the records are placed always in the same position on the films, or if exposure trays are used, upon the trays. These numbers screen the films from the flash of light from above, thus imprinting their images on the films as do the tubes themselves. After the exposure has been given, the radium should be put back in the lead-lined box and removed to the other end of the room while the film is developed. When it is dry, particulars of the tubes can be written in black ink along the top of the contact or face side of the film which already carries the number right way round. Then the numbers of the tubes should be written below each one from the record, and when the film is printed *from the reverse side* the numbering will be correct.

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NOTES

A Proposed Method for Radium Leak Testing by the Use of the Superfluid Properties of Helium

(*Received* 13 *March* 1967)

RADIUM capsules which have excess internal gas pressure and are subjected to the usual stresses of handling, may begin to leak radon through undetectable cracks or fissures. This internal pressure is caused by the inevitable accumulation of helium which accompanies the natural radioactive decay of radium. Therefore, the danger of such leakage is in direct proportion to the container's age and to the quantity of radium it contains.(1)

Sectional diagrams illustrating the construction of typical radium needles and tubes are shown in Fig. 1. Tubes closely resemble needles except that they do not have a sharp point and are larger in diameter.(2)

The viscosity of helium gas which exists in the radium container is 194.1×10^{-6} poise.(3) At 2.18°K which is called the "lambda point", liquid helium has a viscosity one-fifth that of helium in its gaseous state. The liquid helium is commonly called normal fluid.

Below the lambda point, liquid helium becomes a two-fluid model, one of which has the properties of a normal fluid and the other the properties of a superfluid. The superfluid has a very low viscosity which is in the range of 10^{-12} poise.(4) With decrease in temperature below the lambda point, an increasing fraction of the normal fluid is transformed into superfluid until, at absolute zero, the whole liquid is superfluid. The total density of liquid helium, P, is considered as being made up of the normal component, P_n, and that of the superfluid, P_s, so that at all temperatures $P = P_n + P_s$.

It can be seen from Fig. 2 that a 1°K the concentration of normal fluid is negligible.

Normal fluid possesses a volume per second flow inversely proportional to its viscosity, which is the classical behaviour of all fluids. Superfluid flow, on the other hand, is nonclassical and experiments with capillaries of different sizes have shown that this anomalous character becomes even more pronounced with finer caliber tubes. The flow rate of superfluid is dependent only on its temperature. Pressure head and channel length have no effect on the flow rate, even through the narrowest channels. Thus, there will be a noticeable flow of helium through very fine capillaries of fissures at temperatures below the lambda point. This phenomenon has been called "superleak".(5)

Because of the physical properties of liquid helium below the lambda point, a cryostat system can be devised for leak testing and decompression of radium containers. Such a system is shown diagramatically in Fig. 3.

If a radium container is to be leak-tested with the cryostat system in Fig. 3, the container is introduced into the cryostat and liquid nitrogen and helium are transferred into their respective dewars. The system is then evacuated until the radium container is cooled to 1°K. At this temperature helium becomes a superfluid as discussed above, and can, thus, flow through any microscopic cracks in the container through which helium gas or radon could not flow at their normal viscosity. Any leak of helium gas will then be detected by the mass spectrometer. The

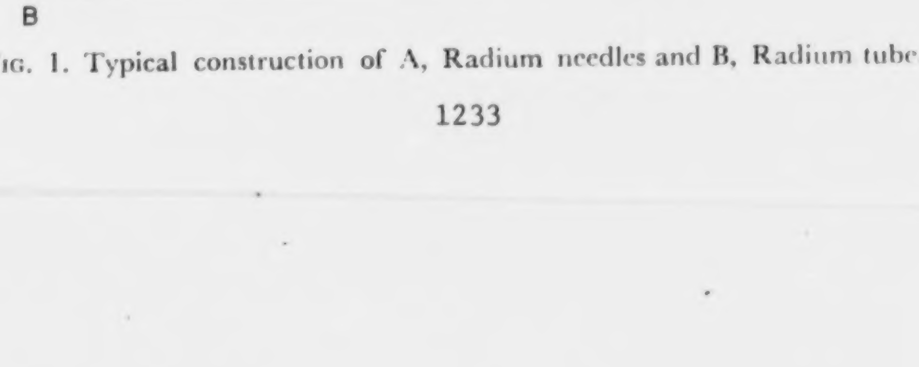

FIG. 1. Typical construction of A, Radium needles and B, Radium tube.

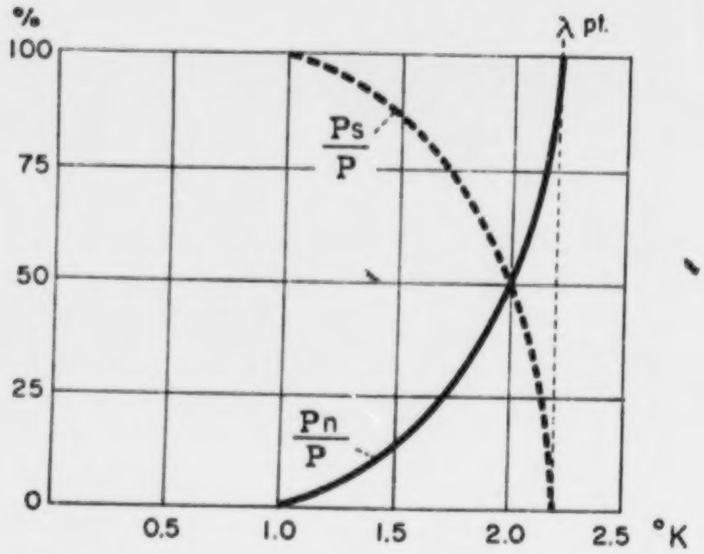

FIG. 2. Relative concentration of superfluid (P_s) and normal fluid (P_n) to total fluid (P) vs. temperature (°K).

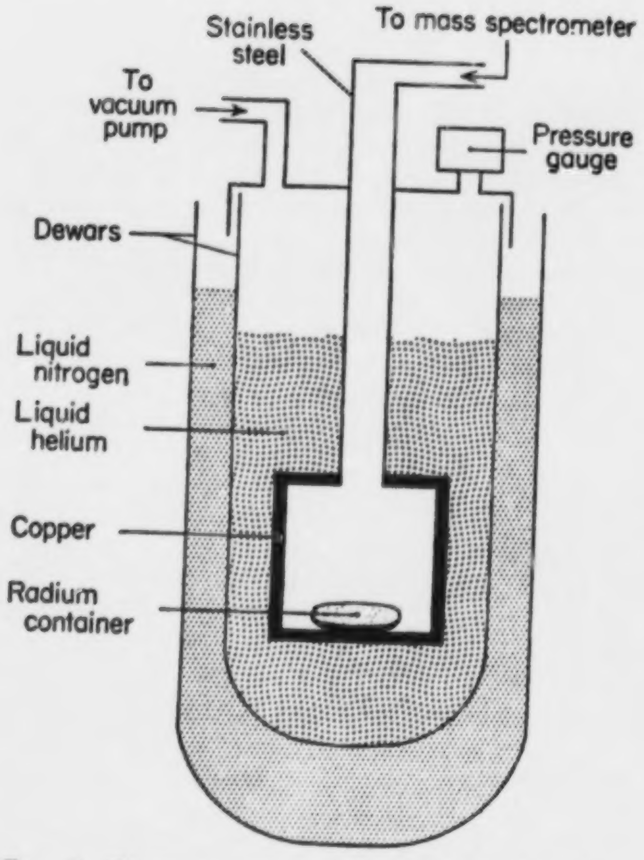

FIG. 3. Diagram of cryostat system for the detection of helium leakage from radium containers.

detection of helium in this system would, therefore, mean that the container is potentially in danger of leaking radon. Decompression of the container could also take place at the time of such testing by removing a sufficient amount of helium.

Caution should be used in testing radium containers in this manner because of potential danger involved since dissimilar metals are used in sealing the radium container, these metals could separate during such cooling and thus might contaminate portions of the cryostat system.

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A NEW METHOD FOR LEAK-TESTING SEALED SOURCES
OF
RADIUM-226 AND THORIUM-228

by

D.J. DWIGHT

ABSTRACT

A simple method has been developed in which radon leaking from radium-226 and thorium-228 sources under vacuum is quantitatively measured. The collection of radon is by dissolution in an organic solvent. Measurement is by liquid scintillation counting, using an internal phosphor, α- and β-particles both being counted with high efficiency. The practical limits of leak detection are:-

Radon-222, 2×10^{-6} μc. per 24 hours
Radon-220, 2×10^{-5} μc. per 24 hours

Natural Radioelements Department,
The Radiochemical Centre,
AMERSHAM.

July 1964

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2. Testing sealed and open sources suspended above scintillant.

3. Testing groups of sources.

4. Checking the emanating rate of an open source.

5-6. Testing the effect on collected activity of the distance between source and collector.

1. INTRODUCTION

Despite the readier availability and lower cost of several artificially produced radioisotopes, radium-226 and to a lesser extent thorium-228 continue to find application in sealed radiation sources. Radium-226 sources are widely used in the treatment of malignant disease, and as γ-radiation standards. Both radium-226 and thorium-228 are used in γ-reference sources and in (α,n) neutron sources.

These sources are tested after manufacture, and at intervals during use, for leakage of emanation. The purpose of such a test is to ensure that no loss of parent activity or of emanation decay products is occurring sufficient to affect significantly the emission of the source, or to cause objectionable contamination of personnel or equipment. The level at which leakage becomes objectionable may vary considerably with the use to which the source is to be put. Recent legislation (1), covering sources in use in the United Kingdom for other than clinical purposes, requires that tests for leakage be periodically performed, and that leak-rates be recorded, the maximum permissible leak for any source to be 0.5 μc. per 24 hours. However for many applications the tolerable leak-rate of a source may be very much less than the legal maximum. In the case of clinical sources, which are frequently handled and stored in large numbers, leak-rates should be of a very low order to avoid exceeding the maximum permissible concentration of radon in air inhaled by the operators (3×10^{-8} μc. per ml.)

The methods in current use for the detection of emanation do not give quantitative results, and so a new method has been developed capable of reproducibly measuring leak-rates, the practical limits of sensitivity being:-

Radon-222	2×10^{-6} μc. per 24 hours
Radon-220	2×10^{-5} μc. per 24 hours

2. PREVIOUS METHODS

Various methods for leak-testing radium-226 and thorium-228 sources have been described.

2.1 The source is placed in a tube part - filled with water, and the pressure in the air-space reduced. The emergence of bubbles indicates a leak (2).

2.2 The source is placed in a closed vessel equipped with two electrodes across which an A.C. potential is applied. The active deposit collected on the electrodes is detected using an α-electroscope (2). In a more recent electrostatic method (3) a 300 volt D.C. supply is used. Collection of active deposit is mainly cathodic since radioactive decay tends to leave the daughter atom positively charged.

2.3 The source is placed in a closed tube with a carbon tablet or a plug of cotton-wool for a period of 24 hours, after which collected activity is detected using an air-ionisation chamber, an end-window Geiger counter or a sodium iodide scintillation counter (2).

2.4 In another variant of the above method a grain of active charcoal is used as a collector (4). Some radon, in addition to active deposit, would in this case be collected.

The bubble-test, 2.1, is useful for detecting flaws in the larger sizes of source container, but is unsuitable for sources with small internal volumes. The results obtained from the emanation methods are difficult to interpret, since the efficiencies of collection and measurement are both uncertain. Because of unknown sensitivity the standard set for passing the test is usually that no detectable activity be found, and consequently, long counting times or slow electroscope discharge rates are involved. Thus, in addition to being merely qualitative, the tests are also tedious, and occupy equipment for long periods.

3. THE PROBLEM

3.1 The collection of emanation

Previous methods for leak-testing have relied mainly on the collection of active deposit. Ideally, the radon itself would be quantitatively trapped. Since radon forms no convenient chemical compounds, practical methods for its collection depend on such processes as condensation, adsorption and dissolution. Of these, dissolution is the simplest, occurring readily at normal temperatures in a number of organic solvents, of which the following are examples:- (5)

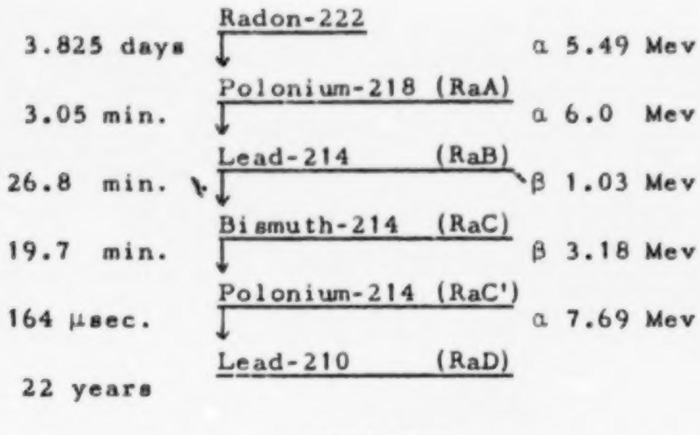
Solvent	K (18°C)
Chloroform	15.08
Benzene	12.82
Toluene	13.24
Xylene	12.75
Ethanol	6.17

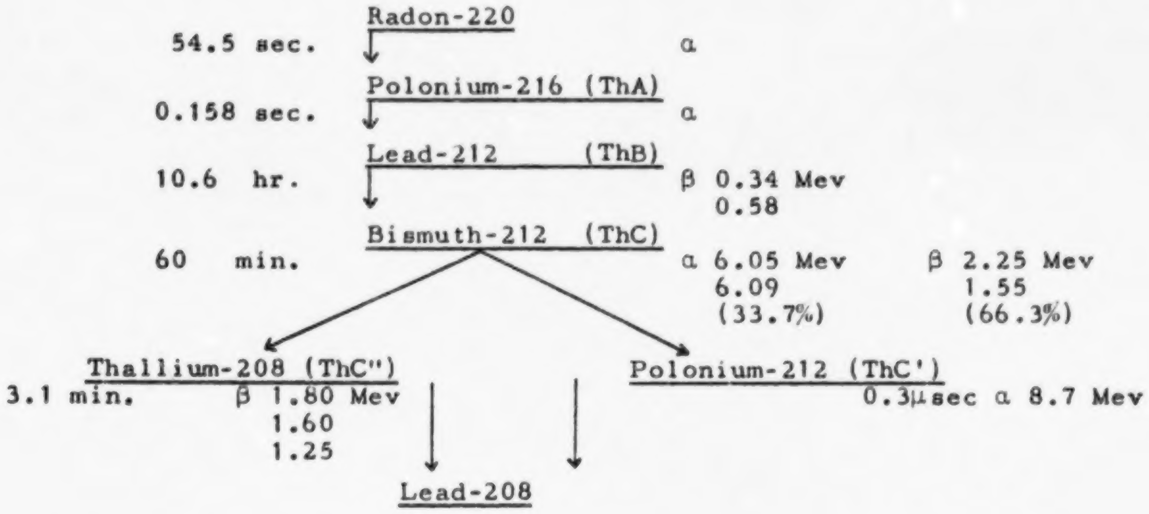
where K is defined as the ratio of the concentration of radon in the liquid phase to that in the gaseous phase.

Thus, over 90% of the radon escaping from a source can be trapped using a suitable solvent.

3.2 The measurement of radon-222 and radon-220

The decay schemes are given below. The minor branches, unimportant in radon measurement, have for simplicity been omitted.


Radon-222
3.825 days
α 5.49 Mev
Polonium-218 (RaA)
3.05 min.
α 6.0 Mev
Lead-214 (RaB)
26.8 min.
β 1.03 Mev
Bismuth-214 (RaC)
19.7 min.
β 3.18 Mev
Polonium-214 (RaC')
164 μsec.
α 7.69 Mev
Lead-210 (RaD)
22 years


Radon-220
54.5 sec.
α
Polonium-216 (ThA)
0.158 sec.
α
Lead-212 (ThB)
10.6 hr.
β 0.34 Mev
0.58
Bismuth-212 (ThC)
60 min.
α 6.05 Mev
6.09
(33.7%)
β 2.25 Mev
1.55
(66.3%)
Thallium-208 (ThC")
3.1 min.
β 1.80 Mev
1.60
1.25
Polonium-212 (ThC')
0.3μsec α 8.7 Mev
Lead-208

Previous methods for detecting activity depended either, on the counting of β- and γ-radiation (with some α-counts if a thin-window counter were used) or on the detection of α-particle ionisation using the relatively insensitive electroscope. Detection efficiencies could be up to about 30% per particle, depending on particle energy and on geometry.

Two methods for counting liquid samples are available:-

3.2.1 Geiger counting

Using an M6 counting tube, the 1.03 Mev and 3.18 Mev β-particles from lead-214 and bismuth-214 could be counted with efficiencies of approximately 1% and 18% respectively. The detection of lead-212, bismuth-212 and thallium-208 would be less efficient overall. Decontamination of the counting tube following high count rates would be a difficulty.

3.2.2 Liquid scintillation counting

This technique involves the addition of the sample to a water-miscible solvent mixture containing a dissolved phosphor.

Detection efficiencies are obtained of 100% for α-particles, and of 80-100% for β-particles according to energy. Solvents commonly used (xylene, dioxan and ethanol) dissolve radon readily, and so the scintillant can be used for direct collection of the activity. No decontamination problem arises since disposable individual glass tubes are used for the samples. This technique, therefore, provides the basis of a particularly suitable method.

4. EXPERIMENTAL WORK

A series of experiments was carried out to test the reliability and to determine the sensitivity of the proposed method, and to evolve a simple technique for routine operation. The initial work was done with radium-226 sources.

4.1 Equipment

4.1.1 Scintillant

Water-miscible scintillant was prepared to the following specification:-

naphthalene (mol. wt. grade)	-	80	grams
2, 5-diphenyloxazole	-	5	"
1, 4-bis- [2-(5 phenyloxazolyl)] benzene	-	0.05	"
xylene (sulphur-free)	-	333	ml.
ethanol	-	"	
dioxan (purified)	-	"	

4.1.2 Test vessels

10 ml. vials with rubber caps.

4.1.3 Vacuum pump

Edwards vacuum pump type RBF1 capable of pumping down to 0.2 atmospheres, with a No.14 hypodermic needle connected via P.V.C. tubing.

4.1.4 Pressure gauge

An Edwards gauge fitted with a No.14 hypodermic needle via narrow-bore P.V.C. tubing.

4.1.5 Counting assembly

Isotope Developments Ltd.	scintillation head	type 2022
	coincidence control unit	type 2032
	fast scaler	type 1009E
	slow scaler	type 1221C

4.2 Experiments

Experiment 1

A number of sources, with a wide range of leak-rates, were submitted for repeated tests, each of 22 hours duration. The sources were immersed in the scintillant as shown in figure 1. The stringency of the test was increased by reducing the pressure in the air-space to 0.2 atmospheres, the self-sealing rubber cap being pierced by the hypodermic needle attached to the pump.

The sources were also tested using the conventional cotton-wool plug collector, the activity being measured with an end-window counter. The results given in Appendix I showed the new test to be much more sensitive than the old, the factor being very approximately 10^4. However, the results obtained with the new test displayed a lack of reproducibility which in some cases was very marked. Experiments were therefore carried out to determine whether these variations were due to fluctuations in the leak-rate of the sources or to unreliability of the test.

Experiment 2

The immersion tests were repeated as above but with the temperature controlled to ± 0.25°C.

Experiment 3

The sources were suspended above the scintillant, using the apparatus shown in figure 2. The temperature was controlled to ± 0.25°C.

Experiment 4

Three unsealed sources were prepared, two of radium barium sulphate and one of radium chloride. Radon from these sources was collected using the apparatus shown in figure 2. Since the volatile alcohol and dioxan might have affected the emanating power of the open sources, xylene only was used as the scintillant solvent. Ambient temperatures and pressures were used.

Experiment 5

The stability of solutions of radon in scintillant was tested by following the decay of the count rates.

The results of these experiments, given in detail in Appendices II to V, showed that:-

(a) The amount of radon collected in the scintillant was not markedly dependent on whether the sources were suspended or immersed.

(b) The results from certain metal-encapsulated sources could vary widely in a series of tests despite control of temperature and pressure.

(c) The collection of radon from open sources was reproducible to ± 10%.

(d) The solutions of radon in scintillant were stable.

It was, therefore, concluded that the method was basically sound, and that the variable results obtained in experiments 2 and 3 were due to fluctuations in the leak-rates of the sources concerned.

5. THE TESTING OF PRODUCTION SOURCES

The new test was applied for three months to the routine production testing of clinical radium needles and tubes. The procedure adopted was as follows:-

(a) Radon was collected over a period of 72 hours from sources placed, singly or in groups of up to five, in tubes of scintillant as shown in figure 3.

(b) After the collection of radon the sources were removed and the activity in each tube was compared with the background count obtained on a blank. A count rate of 1000 per minute above background was set as the tolerance for a source or group of sources. Such a count rate could be measured with adequate precision over a counting period of one minute viz.,

Background (3 x 1 minute counts on a blank)	100 ± 6 cpm
Sample + background (1 minute)	1100 ± 33 cpm
∴ Sample count rate	1000 ± 34 cpm standard deviation

(c) Groups which failed the test were split and the individuals retested separately.

Sources passing the test were submitted for the conventional test.

5.1 Results

262 groups of sources, containing 787 individual sources, were tested. 80% of the test results were within the set tolerance, 55% being above background by a scarcely significant margin. After individual tests on groups which failed, it was found that 92% of the sources were satisfactory. An analysis of the results is given in Appendix VI.

All the sources passing the new test were successful in the conventional test, so that there was no evidence over the period of the test programme of the new method failing to detect any leaks.

6. DETERMINATION OF THE SENSITIVITY OF THE NEW TEST

6.1 Counting of α-particles

A polonium-210 solution was assayed by counting dried aliquots in an argon-flow proportional counter. A plutonium-239 source calibrated in a 2π methane-flow proportional counter was used as a standard. Aliquots from this solution were added to samples of scintillant and the activity measured in the I.D.L. 6012 counter.

A counting efficiency of 100% was indicated. Details of the results and the operating conditions of the counter are given in Appendix VII.

6.2 Counting of radium-226 in equilibrium with its decay products

Four glass capillary tubes of radium chloride, measured using a sodium iodide crystal counter to ± 4% maximum error, were used to prepare standard solutions of radium-226. Aliquots from these solutions were added to 4 ml. volumes of scintillant in glass ampoules. The ampoules were flame-sealed and stored for 35 days in a water-bath at 23°C to allow attainment of radioactive and partition equilibria. Twenty-four ampoules, giving count rates between 5×10^4 and 5×10^5 per minute, were given three measurements each, with an interval of one week between measurements. The contribution of the radium-226 α-particle to the count rate was subtracted, and the detection efficiency of the system for radon-222 calculated to be 0.909.

Full details of the results and the operating conditions of the counter are given in Appendix VIII.

6.3 The rate of attainment of partition equilibrium

6.3.1 Radon was injected into a glass ampoule half full of scintillant. The ampoule was flame-sealed and placed in a water-bath at 23°C. At intervals the ampoule was transferred to a scintillation counter and the count rate measured. Four such tests indicated that partition equilibrium was reached in three hours.

6.3.2 An ampoule was half filled with a solution of radon in scintillant and left for a week for equilibrium to be reached. The seal was then broken, the radon expelled from the air-space by an air-jet, and the ampoule resealed. The count rate was measured at intervals. As before, equilibrium was reached after three hours.

The results of these experiments are given in graph I in Appendix IX.

It follows that in accurate leak-rate measurements certain corrections should be made:-

6.3.3 When the collection of radon is terminated, that fraction of the radon which has entered the system during the previous three hours is not in partition equilibrium. In the case of sources suspended over scintillant graph Ia applies and indicates a positive correction. In the case of immersed sources graph Ib applies and indicates a negative correction. For 24 hour collection periods the corrections, assuming immediate counting, are + 2.2% and -2.8% respectively, and for collection periods of 48 hours or more, are less than 1%.

6.3.4 When the source is removed at the end of the collection period the radon in the air-space is lost from the system by diffusion and turbulence. A fresh partition equilibrium forms, the rate being shown in graph Ib. The correction to be applied to the count rate obtained at a known time after disturbing the original equilibrium can be obtained from graph II in Appendix IX.

7. CHECKING THE ACCURACY OF THE NEW METHOD

7.1 Measurements on an open source

7.1.1 The emanating rate of a radium barium sulphate source was found using the apparatus shown in figure 2. Sufficient moisture was added to the system to saturate the air. Pressure was ambient, but the temperature was controlled at 23°C ± 0.25°C.

7.1.2 The emanating rate of the source was then measured as follows. The source was sealed in a tube as shown in figure 4, and radon allowed to accumulate for several days. Pressure was ambient, but the temperature was controlled at 23°C ± 0.25°C. The tube was then flame-sealed at the centre, "X", and the radon in section A measured using a sodium iodide crystal counter. A radium chloride capillary, of similar physical size and of approximately the same activity, was used as a standard. Both sections of the tube were then cracked open, and their internal volumes and the volume of the source and water-tube determined, so that the total amount of radon emanated could be calculated. The results are given in Appendix X.

8. THE LEAK-TESTING OF THORIUM-228 SOURCES

In applying the new method to the testing of thorium-228 sources account had to be taken, at both collection and measurement stages, of the short half-life of the radon-220 (54.5 sec.).

8.1 The collection of emanation

The half-life of radon-220 is sufficiently short for significant decay to occur before the gas has diffused uniformly throughout the air-space in the leak-test tube. Thus the activity collected varies with the distance separating the source and collector. The effect was demonstrated by the following experiments.

8.1.1 Emanation from two thorium-228 sources and one radium-226 source was collected on short lengths of thin-walled polythene tubing placed at varying distances from the sources in the leak-test tubes. The apparatus is shown in figure 5. After 72 hours the count rate on each piece of tubing was measured using an end-window counter. A marked fall in count rate with distance from the source occurred with thorium-228, whereas no such effect was observed with radium-226.

8.1.2 Emanation was collected from a thorium-228 source using scintillant at varying distances from the source. The apparatus is shown in figure 6. A sharp fall in count rate with distance from the source indicated that quantitative collection of activity from thorium-228 sources could only be achieved by immersion in the scintillant.

The results of these experiments are given in Appendix XI.

8.2 The measurement of emanation

Because of the rapid decay of radon-220 the measurement of the collected activity depends upon the formation of stable solutions of lead-212 and bismuth-212, the 10.6 hour and 60 minute daughters. It was found that the addition of a few μgm. of lead and bismuth carriers, as the nitrates in 0.1 normal nitric acid, resulted in stable solutions of activity in scintillant. In the absence of carriers, instability sometimes though not always occurred. The results of this work are given in Appendix XII.

The effective number of particles counted is three, viz.

100%	β	(Pb-212)	0.34 Mev 0.58	80% 12%
66.3%	β	(Bi-212)	2.25, 1.55 Mev	
33.7%	α	"		
66.3%	α	(Po-212)		
33.7%	β	(Tl-208)	1.80, 1.62, 1.25 Mev	

The counting efficiency for α-particles has been shown to be 100%. (Appendix VII). Experience in the Analytical Department of The Radiochemical Centre with the measurement of a number of β-emitting isotopes indicates that the bismuth-212 and thallium-208 β-particles would be counted with 100%, and the lead-212 β-particles with approximately 90%, efficiency. Thus the overall counting efficiency is expected to be 96%.

8.3 The sensitivity of radon-220 detection

The immersion of sources in scintillant results in effectively 100% trapping of activity since partition between the liquid and gaseous phases within the life-time of radon-220 is insignificant, as graph Ib in Appendix IX shows. This point has been substantiated by direct experiment. Thus the overall detection efficiency of radon-220 in the immersion-testing of sources can be expected to be approximately 0.96.

8.4 Leak-testing metal-encapsulated thorium-228 sources

Four sources were tested by immersion in scintillant and then by the cotton-wool method, the plug of wool being placed approximately 1 cm. from the source.

The results, given in Appendix XIII, show the improved sensitivity of the new method.

9. CONCLUSION

The collection of emanation in liquid scintillant, followed by its measurement by scintillation counting, is a simple, reliable and sensitive leakage test for sealed sources of radium-226 and thorium-228.

The efficiencies of collection and measurement being known, leak-rates under the conditions of the test may be calculated.

Radium-226 sources may be either immersed in the scintillant or suspended above it. In practice immersion is more convenient, although it seems reasonable to suppose that results obtained on the suspended source might more closely represent the leak-rate to be expected in free air.

Thorium-228 sources must be immersed in the scintillant because of the short half-life of the radon-220.

10. SUMMARY OF NEW TEST PROCEDURE

(a) Place source in a glasstube half full of liquid scintillant to which a few μgm. of lead and bismuth carriers have been added.

(b) Close tube with a rubber cap and pump the air down to low pressure using a hypodermic needle to pierce the cap.

(c) Allow several days (at least three) for collection of emanation, so that the leak-rate of an erratically leaking source may be reliably sampled. Store in the dark to avoid photoluminescence.

(d) Remove source and transfer stoppered tube to scintillation counter, avoiding exposure to direct sunlight. Count against a blank.

(e) If the leak-rate of the source is required, calculate using the factors:-

0.91 for radon-222

Note: The count rate should be corrected for partition adjustment. (See paras. 6.3.3, 6.3.4, and Appendix IX.

0.96 for radon-220

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APPENDIX I

RESULTS OF LEAK-TESTS PERFORMED UNDER VARIOUS CONDITIONS ON A GROUP OF SOURCES

Sources immersed in scintillant at ambient temperature

Source number				cpm per 24 hours			
1	2	3	4	5	6	7	8
20854	4479	97114	22145	1.16×10^5	306	2.0×10^6	241
19945	5524	48890	25436	21754	321	4.5	407
21310	4909	38380	5165	94785	286	2.8	274
15887	4585	51426	30848	3.27×10^5	243	7.6	262
26886	5020	41900	14517	2.7 "	308	2.2	454
38721	4160	56892	15615	1.44 "	260	3.2	338
			Mean counts				
2.4×10^4	4.8×10^3	5.6×10^4	2.6×10^4	1.6×10^5	287	3.7×10^6	330

Sources tested by the cotton-wool method

Source number				cpm per 24 hours			
1	2	3	4	5	6	7	8
2.5	NS	12	1.4	0.9	NS	909	NS
NS	NS	4.5	1.3	0.8	NS	742	NS
NS	NS	5.5	2.0	7.5	NS	253	NS
3	NS	10	2.4	3.4	NS	5017	NS
NS	NS	6	NS	2.4	NS	1325	NS
5		8.5					
			Mean counts				
1.8	NS	7.7	1.4	3.0	NS	1.6×10^3	NS
		Ratio of sensitivity, new test : old					
1.3×10^4	-	7.3×10^3	1.8×10^4	5.3×10^4	-	2.3×10^3	-

Notes:
1. NS indicates count not significantly above background.
2. Count-rates obtained in tests on source 7 were probably too high for the counter to record accurately.

APPENDIX II

Sources immersed in scintillant. Temperature 25°C ± 0.25°C

Source number		cpm per 24 hours	
1	2	4	7
19264 18154 11730 20000	3156 3351 3400 5096	19383 33458 5474 11462	2.8×10^6 4.5 " 5.5 " 10 "

APPENDIX III

Sources suspended over scintillant. Temperature 25°C ± 0.25°C

Source number				cpm per 24 hours		
1	2	4	5	6	7	8
26505 22677 26464 19224	4900 4753 5095 4454	11140 17193 7642 12504	1.01×10^5 0.76 " 1.55 " 21703	215 243 204 193	9.8×10^6 7.6 " 6.7 " 9.6 "	222 251 175 177

APPENDIX IV

The collection of radon from open sources cpm per 24 hours

Radium barium sulphate source 10	Radium barium sulphate source 11	radium chloride source 12
6.10×10^5 5.07 5.38 5.86 5.70 5.66 5.64 5.26 5.20 5.45	9.55×10^5 9.12 9.49 9.24 9.51 10.1 10.5 10.3 10.0 9.35	3.65×10^4 3.48 3.70 3.72 3.68 3.58 3.60 3.65 3.60 3.72
	Mean counts:	
5.53×10^5 ± 10%	9.71×10^5 ± 8%	3.63×10^4 ± 4%

APPENDIX V

The decay of samples

Sample	Decay (days)	Counts/min. observed	Counts/min. calculated
1	0	160457	-
	1	137951	1.33×10^5
	3	94094	0.93
	6	52483	0.54
	10	24266	0.26
2	0	6628	-
	4	3076	3180
	11	680	860
3	0	97114	-
	4	44443	4.60×10^4
	11	11203	1.26
4	0	2349	-
	1	1859	1950
	3	1327	1360
5	0	809	-
	1	716	675
	3	470	470
6	0	23742	-
	1	20115	1.98×10^4
	3	13320	1.38

APPENDIX VI

Testing of production sources : analysis of results

Range cpm	<50	51 to 100	101 to 200	201 to 400	401 to 600	601 to 1000	1001 to 1500	1501 to 3000	3001 to 5000	5001 to 10^4	$>10^4$
Groups %	55.5	9.3	4.2	5.0	3.5	2.3	1.9	4.6	2.3	1.9	9.5
Passes 80%							Failures 20%				

APPENDIX VII

The response of the I.D.L. Tritium scintillation counter type 6012 to α-particles

Polonium-210 solution assay 0.300 mc./gm. Argon-flow counter standardised using a plutonium-239 source calibrated by 2π counting.

	Calculated	Efficiency
Sample 1:-		
52.5 mg solution gave 35816 cpm	3.50×10^4 dpm	102%
Sample 2:-		
22.3 mg solution gave 15018 cpm	1.49×10^4 dpm	101%

E.H.T. 1800 volts per channel. Bias 10 volts. Gate "OUT".
Dead-time 4 μsec. each channel. Resolving time 0.1 μsec.

APPENDIX VIII

The counting of radium in equilibrium with its decay products

I. 11.37 μg radium in 51 gm. solution

Tube	mg soln.	meast. 1	meast. 2	meast. 3	calc. count	meast.* / calc.
1	15.8	4.217×10^4	4.214×10^4	4.210×10^4	4.70×10^4	0.909
2	17.0	4.527	4.537	4.540	5.05	0.910
3	18.9	5.007	4.990	4.993	5.61	0.904
4	18.8	5.014	5.008	5.049	5.58	0.912
5	18.3	4.873	4.856	4.881	5.44	0.908
6	15.8	4.235	4.232	4.196	4.69	0.912

II. 107.2 μg radium in 50 gm. solution

Tube	mg soln.	meast. 1	meast. 2	meast. 3	calc. count	meast.* / calc.
7	20.9	5.038×10^5	5.105×10^5	5.093×10^5	5.96×10^5	0.940
8	21.6	5.223	5.331	5.330	6.17	0.948
9	21.6	5.206	5.284	5.224	6.17	0.937
10	21.7	5.192	5.268	5.260	6.20	0.931
11	21.5	5.281	5.321	5.311	6.19	0.952
12	21.5	5.221	5.249	5.245	6.15	0.941

III. 96.5 μg radium in 250 gm. solution

Tube	mg soln.	meast. 1	meast. 2	meast. 3	calc. count	meast.* / calc.
13	22.5	1.056×10^5	1.059×10^5	1.061×10^5	1.16×10^5	0.932
14	22.0	1.027	1.037	1.029	1.13	0.930
15	47.2	2.162	2.182	2.199	2.43	0.945
16	42.9	1.990	1.991	2.003	2.21	0.945
17	66.1	2.945	2.971	2.978	3.40	0.935
18	23.3	1.040	1.046	1.053	1.20	0.900

IV. 12.1 μg radium in 50 gm. solution

Tube	mg soln.	meast. 1	meast. 2	meast. 3	calc. count	meast.* / calc.
19	47.1	1.324×10^5	1.333×10^5	1.328×10^5	1.52×10^5	0.900
20	40.2	1.132	1.148	1.150	1.29	0.917
21	64.7	1.831	1.841	1.836	2.06	0.928
22	44.8	1.251	1.280	1.277	1.44	0.910
23	45.0	1.269	1,282	1.278	1.45	0.920
24	72.2	2.035	2.063	2.050	2.33	0.923

Grand mean $\frac{\text{meast.}}{\text{calc.}} = 0.924.$

Let detection efficiency for radon be e.
Efficiency for the radium α-particle = 1.

$$\therefore \quad \frac{5e + 1}{6} = 0.924$$

$$\therefore \quad \underline{e = 0.909}$$

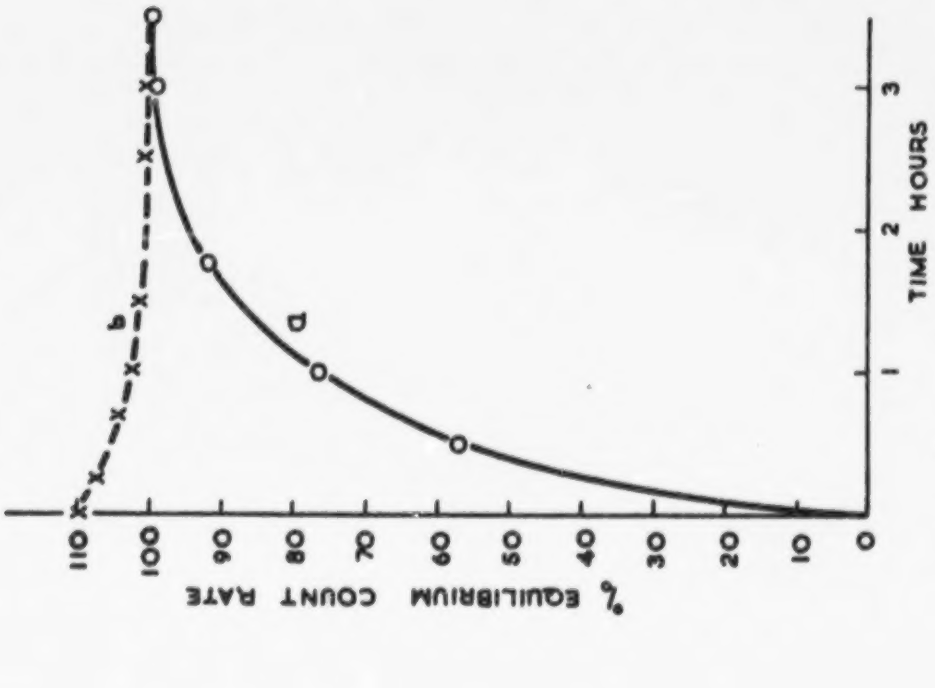
*Mean count corrected for coincidence losses using formula:-

$$\text{Corrected count} = \frac{C - Tr.A^2}{1 - AT} \times \frac{1}{1 - A(T + 2Tr) - CT} \quad \ldots\ldots\ldots\ldots \quad (6)$$

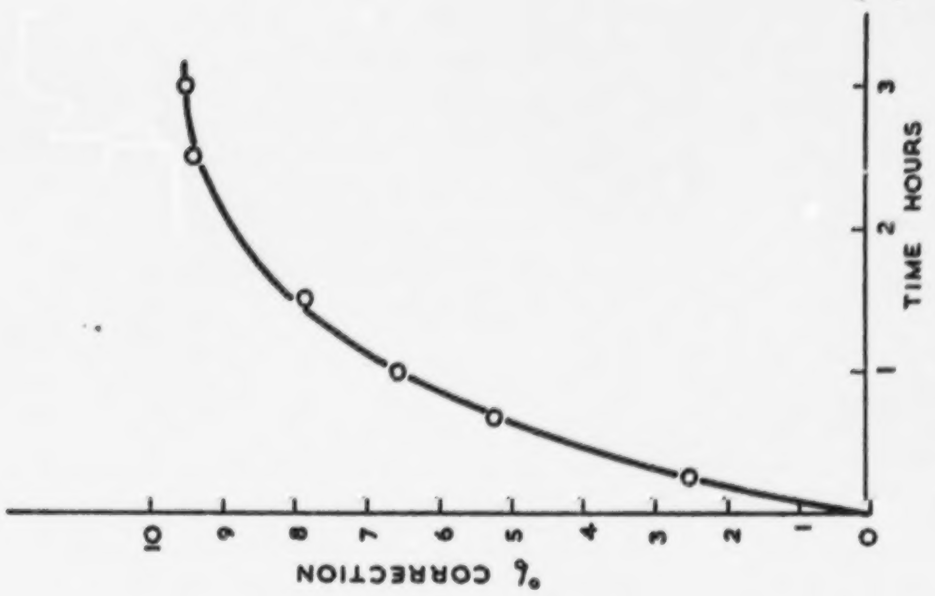
where: A = count in either channel, Tr (resolving time) = 0.1 μs
C = count observed T (dead time) =.4 μs

APPENDIX IX

Rate of attainment of partition equilibrium



I a. RADON INITIALLY IN GASEOUS PHASE.
b. RADON INITIALLY IN LIQUID PHASE.



II CORRECTION TO COUNT RATE FOR ADJUSTMENT OF PARTITION FOLLOWING DISTURBANCE AT 0 HOURS.

APPENDIX X

Measurements on the emanating rate of an open source

I. Scintillation measurement (7.1.1)

Collecting time hr.	Count cpm.	Radon µc./24 hr.	
24	7.06×10^5	0.070	
23	6.76	0.070	
24	7.41	0.073	Mean
20	5.59	0.066	0.070 µc./24 hr.

Note: Counts corrected for coincidence losses and for partition adjustment.

II. Check measurement (7.1.2)

Sample	Collection time hrs.	Meast. µc.	Vol. A ml.	Vol. B ml.	Vol. source ml.	Total µc./24 hr.
1	120	0.116	0.410	0.450	0.059	0.063
2	71	0.084	0.380	0.410	0.059	0.065
3	71	0.0845	0.307	0.353	0.059	0.067
4	69	0.077	0.344	0.408	0.059	0.064
					Mean	0.065 µc./24 hr.

APPENDIX XI

The effect of distance from source on collected activity

I. Polythene tube collectors count rate vs. distance

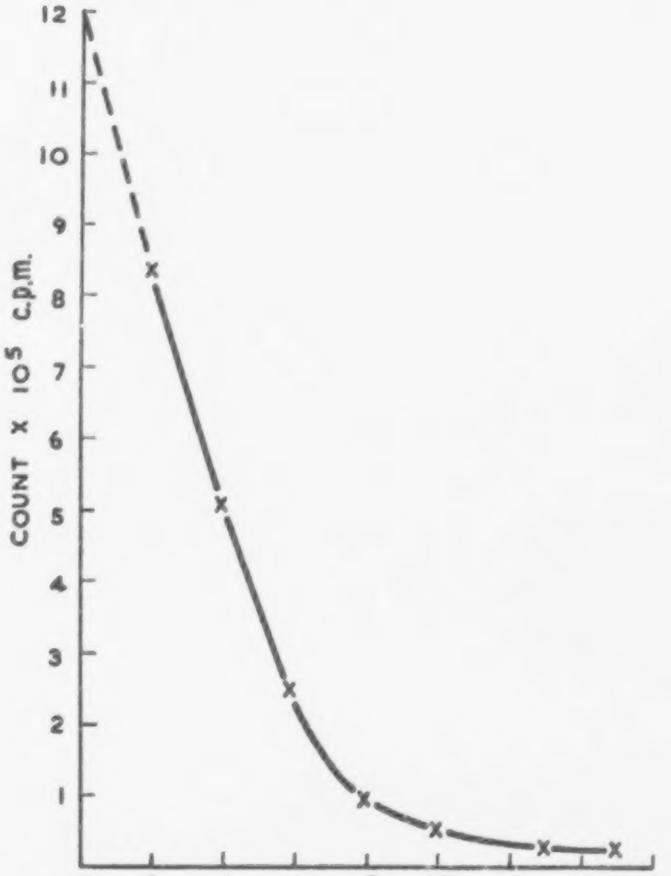
Distance mm.	Thorium-228.1	Thorium-228.2	Radium-226
35	6175 cpm	7017 cpm	737 cpm
65	3976	3283	870
95	1269	1105	742
125	623	799	813

where distance is measured from source to centre of collector

II. Scintillant collector

d cm.	count cpm.
2	8.40×10^5
4	5.19
6	2.52
8	0.955
10	0.530
13	0.270
15	0.224

Count extrapolated
to d = 0 : 1.2×10^6 cpm



APPENDIX XII

The stability of solutions of radon-220 decay products

Time	Sample 1	Sample 2	Sample 3	Sample 4
0	1.74×10^5	1.63×10^5	2.8×10^5	2.52×10^5
5 min.	1.20×10^5	1.60	-	-
10 min.	0.78×10^5	1.59	-	-
15 min.	-	1.58	-	-
25 min.	-	1.56	-	-
24 hours	-	-	0.52	0.51
			calculated 0.59	calculated 0.51

Notes: Sample 1 was in carrier-free solution.
Samples 2-4 contained lead and bismuth carriers.

APPENDIX XIII

Leak-testing metal-encapsulated thorium-228 sources

I. Sources immersed in scintillant

Source number		cpm per 24 hr.	
1	2	3	4
17800	4713	5284	1.12×10^5
19100	3540	3533	0.67
14551	4206	1871	0.66
15443	6663	1977	0.70
	Mean counts		
1.7×10^4	4.8×10^3	3.1×10^3	8.4×10^4

II. Cotton-wool test. Collector 1 cm. from source

Source number		cpm per 24 hr.	
1	2	3	4
8	11	7	20
7	17	10	7
6	9	10	NS
3	6	NS	NS
	Mean counts		
6	11	7	7

Note: NS indicates count not significantly above background.

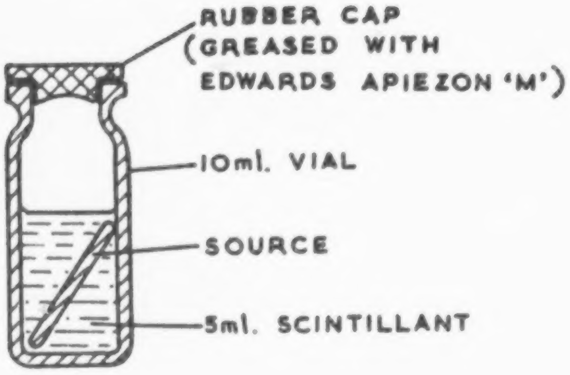

RUBBER CAP (GREASED WITH EDWARDS APIEZON 'M')
10ml. VIAL
SOURCE
5ml. SCINTILLANT

FIG. 1.

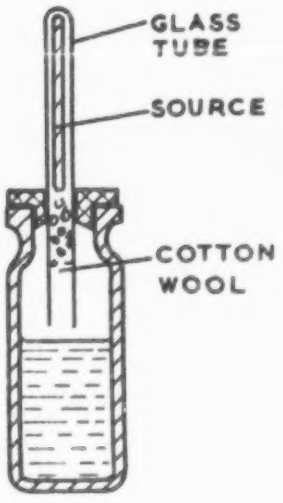

GLASS TUBE
SOURCE
COTTON WOOL

FIG. 2.

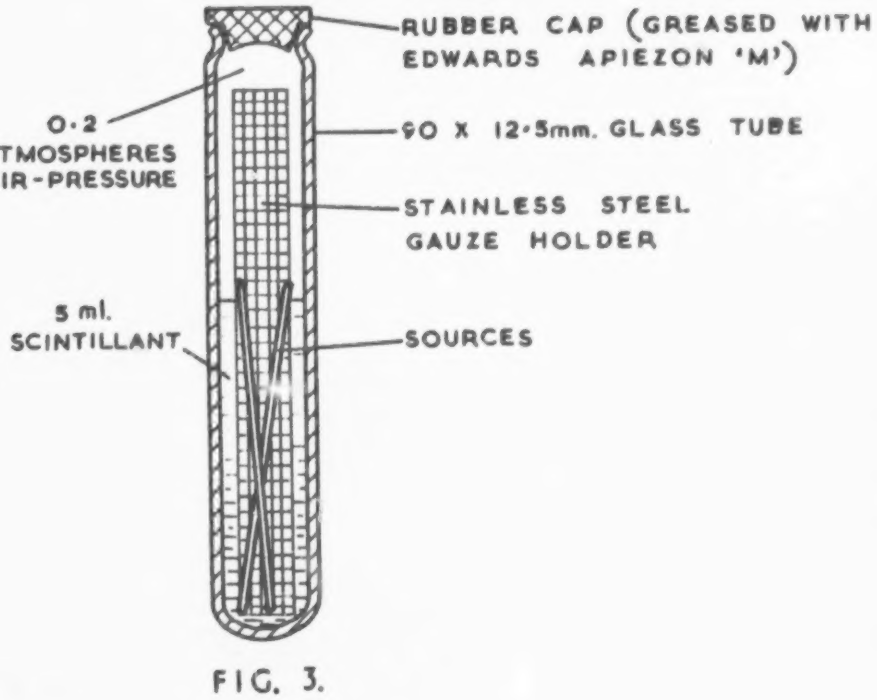

RUBBER CAP (GREASED WITH EDWARDS APIEZON 'M')
0·2 ATMOSPHERES AIR-PRESSURE
90 X 12·5mm. GLASS TUBE
STAINLESS STEEL GAUZE HOLDER
5 ml. SCINTILLANT
SOURCES

FIG. 3.

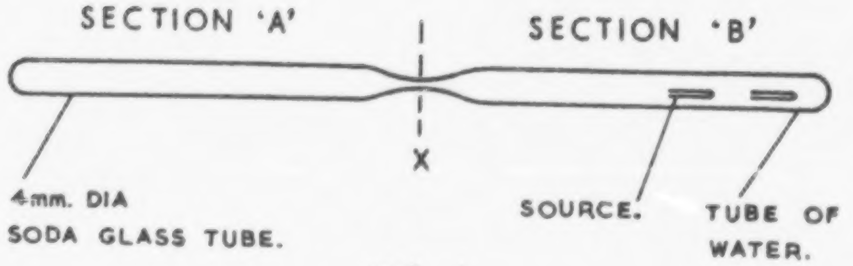

SECTION 'A'
SECTION 'B'
X
4mm. DIA
SODA GLASS TUBE.
SOURCE.
TUBE OF
WATER.

FIG. 4.

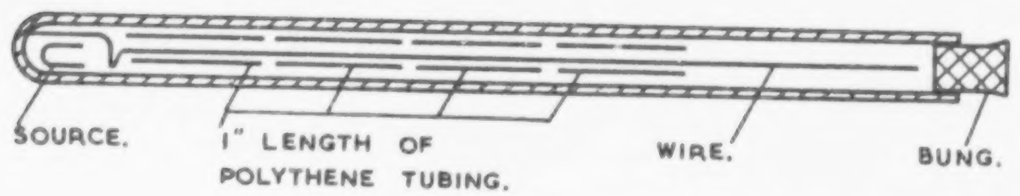

SOURCE.
1" LENGTH OF
POLYTHENE TUBING.
WIRE.
BUNG.

FIG. 5.

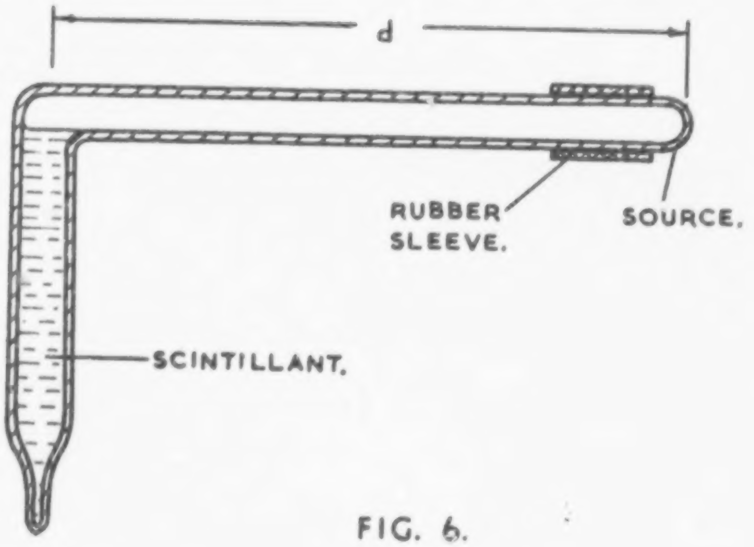

d
RUBBER
SLEEVE.
SOURCE.
SCINTILLANT.

FIG. 6.

UNCLASSIFIED
(Approved for Sale)

RCC-R 176.
(Addendum)

A NEW METHOD FOR LEAK - TESTING SEALED SOURCES
OF
RADIUM-226 AND THORIUM-228
(ADDENDUM)

D. J. DWIGHT

This addendum has been issued to clarify the quantitative interpretation of results obtained from the leak-testing of sealed sources of radium-226 and thorium-228 by the method described in report R.C.C. R.176.

Natural Radioelements Department,
The Radiochemical Centre,
Amersham,
Bucks.

May 1965

HL65/2968
AKC

THE CALCULATION OF RADON LEAK-RATES

The results obtained from radon leak-tests may be expressed as follows:-

1. The radon present at the end of a given period in a sealed enclosure containing a leaking source.
2. The active deposit accumulated in the vicinity of the source as a result of a leak.
3. The total quantity of radon leaked into the sealed enclosure during a given period.

These quantities may be calculated as follows:-

Let the source be enclosed with the scintillant for a time t, and let the count-rate in the scintillant after time t be C_t per minute above background.

In the case of radon-222 detection:- C_t is the total count-rate from radon-222 (half-life 3.825 days) and the four short-lived daughters in equilibrium (Po-218, Pb-214, Bi-214, Po-214). The activity of Pb-210 is negligible. Thus the daughters contribute four disintegrations per radon-222 disintegration. Let the mean detection coefficient for the five disintegrations be 0.91 (see R.176 page 7) Let t be at least 24 hours.

Then, within the limits of experimental accuracy,

total activity after time t

$$= \frac{C_t}{0.91 \times 3.7\times10^4 \times 60} \ \mu c. \quad ....(A)$$

radon activity after time t

$$= \frac{C_t}{0.91 \times 5 \times 3.7\times10^4 \times 60} \ \mu c. \quad ......(B)$$

radon daughter activity after time t

$$= \frac{4\times C_t}{0.91 \times 5 \times 3.7\times10^4 \times 60} \ \mu c. \quad ......(C)$$

Assume that the source leaks radon-222 into the sealed enclosure at a constant rate of Z atoms per second, and let N be the number present after time t.

The rate of accumulation of radon-222 in the sealed enclosure,

$$\frac{dN}{dt} = \text{rate of leakage} - \text{rate of disintegration}$$

$$= Z - \lambda_1 N \quad ....(D)$$

where λ_1 is the decay constant of radon-222

Solving equation D gives the following expression for N,

$$N = \frac{Z}{\lambda_1}\,(1 - e^{-\lambda_1 t}) \qquad \text{......................(E)}$$

Hence the number of radon-222 disintegrations per second at time t will be,

$$\lambda_1 N = Z\,(1 - e^{-\lambda_1 t}) \quad = \quad \frac{C_t}{0.91 \times 5 \times 60}$$

$$\therefore \quad Z \quad = \quad \frac{C_t}{0.91 \times 5 \times 60(1 - e^{-\lambda_1 t})} \qquad \text{atoms per second}$$

and the radon-222 activity leaking per second,

$$\lambda_1 Z \quad = \quad \frac{\lambda_1 \times C_t}{0.91 \times 5 \times 60(1 - e^{-\lambda_1 t})} \qquad \text{d.p.s.}$$

$$= \quad \frac{\lambda_1 \times C_t}{0.91 \times 5 \times 60(1 - e^{-\lambda_1 t}) \times 3.7\text{x}10^4} \qquad \mu\text{c.}$$

Expressing λ_1 as $\frac{0.693}{3.825}$ day^{-1}, the radon-222 leak-rate per 24 hours becomes

$$\frac{0.693 \times C_t}{0.91 \times 5 \times 60(1 - e^{-\lambda_1 t}) \times 3.7\text{x}10^4 \times 3.825} \qquad \mu\text{c.}$$

$$= \quad \frac{C_t \times 1.80 \times 10^{-8}}{(1 - e^{-\lambda_1 t})} \qquad \mu\text{c.} \qquad \text{..............(F)}$$

Limit of detection. In practice the lowest count-rate readily detectable above background is approximately 100 c.p.m.

Then, from eqn. B,

$$\underline{\text{minimum detectable radon-222}} \quad = \quad \frac{100}{0.91 \times 5 \times 3.7\text{x}10^4 \times 60} \qquad \mu\text{c.}$$

$$= \quad \underline{10^{-5}\ \mu\text{c}}.$$

Let the above quantity be that present in the sealed enclosure after a period of 7 days, then, radon-222 accumulated in a 24 hour period $= \frac{10^{-5} \times 0.166}{0.72}\ \mu c.$

$= \underline{2.3 \times 10^{-6}\ \mu c}.$

where 0.166, 0.72 are radon-222 growth fractions
after 1 day and 7 days respectively.

This quantity is that quoted on page 1 of R.176 as the limit of leak detection. The total quantity leaked in a 24 hour period is slightly greater: from eqn. F, radon-222 leak rate $= \frac{100 \times 1.80 \times 10^{-8}}{(1 - 0.28)}\ \mu c./24hr.$

$= \underline{2.5 \times 10^{-6}\ \mu c./24\ hrs}.$

In the case of radon-222 detection: Assume that the Ra-224 (ThX) has been allowed to grow to equilibrium with the thorium-228 before commencement of the test. Since radon-220 is short-lived, it quickly builds up in the sealed enclosure until the disintegration rate equals the number of atoms emerging from the source per second (Z). However, the radon-220 and its first daughter Po-216 decay to negligible levels before measurement. Hence the observed count C_t is the total count from the Pb-212 and its short-lived daughters in equilibrium, viz. Bi-212, Tl-208, Po-212. (Details of branched decay and half-lives are given on page 3 of R.176).

It is necessary, therefore, to calculate the radon-220 activity from the measured activity of Pb-212.

The effective number of particles in the decay chain is three (see R.176 page 9). However, with a counter dead-time long compared with the 0.3 μsecond half-life of Po-212, the disintegration of the Po-212 will not be resolved from that of its parent Bi-212. Hence, the number of counts per radon-220 disintegration will be only 2.3. The mean detection coefficient is 0.96 (see R.176 page 9).

Thus, the total radon-220 daughter activity after time t

$$= \frac{C_t}{0.96 \times 3.7 \times 10^4 \times 60}\ \mu c. \ \ldots\ldots\ldots\ldots\ldots (C')$$

and hence, the Pb-212 activity after time t

$$\frac{C_t}{0.96 \times 2.3 \times 3.7 \times 10^4 \times 60}\ \mu c.$$

Applying eqn. E to the accumulation of Pb-212 in the sealed enclosure, the number (N) of atoms present after time t is given by,

$$N = \frac{Z}{\lambda_2}(1 - e^{-\lambda_2 t}) \quad \text{where } \lambda_2 \text{ is the decay constant of Pb-212}$$

Hence the activity of Pb-212 at time t

$$= \lambda_2 N = Z(1 - e^{-\lambda_2 t}) = \frac{C_t}{0.96 \times 2.3 \times 60} \quad \text{d.p.s.}$$

$$\therefore Z = \frac{C_t}{0.96 \times 2.3 \times 60(1 - e^{-\lambda_2 t})} \quad \text{atoms per second}$$

Radon-220 leak rate per second = Z atoms

$$= \lambda_3 Z \quad \text{d.p.s.} \quad \text{where } \lambda_3 \text{ is the decay constant of radon-220}$$

$$= \frac{\lambda_3 C_t}{0.96 \times 2.3 \times 60(1 - e^{-\lambda_2 t})} \quad \text{d.p.s.}$$

Expressing λ_3 as $\frac{0.693 \times 60 \times 60 \times 24}{54.5}$ day^{-1}

Radon-220 leak-rate per 24 hours

$$= \frac{C_t \times 0.693 \times 60 \times 60 \times 24}{0.96 \times 2.3 \times 3.7\times10^4 \times 60(1 - e^{-\lambda_2 t}) \times 54.5} \quad \mu\text{c.}$$

$$= \frac{C_t \times 2.2 \times 10^{-4}}{(1 - e^{-\lambda_2 t})} \quad \mu\text{c.} \; \ldots\ldots\ldots\ldots (F')$$

For t > 48 hours, when the Pb-212 has grown to within 5% of the Radon-220, within the limits of experimental error,

Radon-220 leak-rate per 24 hours $= C_t \times 2.2 \times 10^{-4}$ μc. ......(F'')

$$\underline{\text{Radon-220 present after time } t} = \frac{C_t}{0.96 \times 2.3 \times 3.7\times10^4 \times 60} \ \mu\text{c.} \quad ..(B')$$

Limit of detection. In practice the lowest count-rate readily detectable above background is approximately 100 c.p.m.

Then, from eqn. B'

$$\text{minimum detectable radon-220} = \frac{100}{0.96 \times 2.3 \times 3.7\times10^4 \times 60} \ \mu\text{c.}$$

$$= \underline{2.1 \times 10^{-5} \ \mu\text{c}}.$$

This quantity is that quoted on page 1 of R.176 as the limit of leak detection. The total quantity leaked in any 24 hour period is very much greater than that present at the end of the period:

For t = 3 days, from eqn. F''

$$\underline{\text{Radon-220 leak-rate}} = 100 \times 2.2 \times 10^{-4} \ \mu\text{c./24 hours}$$

$$= \underline{2.2 \times 10^{-2} \ \mu\text{c./24 hours}}$$

Acknowledgements The author wishes to thank Mr. G.H.C. Dancer and Mr. H. Sheard for their help in the preparation of this addendum.

PRECEDING
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Use of Autoradiographs to Detect Defects in Radium Needles and Tubes and Inequalities in the Distribution of the Radium[1]

JOHN H. FREED, M.D., EUGENE P. PENDERGRASS, M.D., and HENRY RAUFER[2]

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IT IS NOW WELL RECOGNIZED by most radiation therapists that careful preliminary planning and dosage calculations are essential to good radium therapy. Dosage systems have been published by Quimby (5) and Paterson and Parker (3, 4), which provide rules for the proper distribution of radium sources, and tables and charts for calculating the dosage. These dosage systems are based for the most part on the assumption that the radium sources used are free of defects and that the radium is uniformly distributed along the active length of the source. Badly packed needles, damaged containers permitting leakage of the radium, and faulty distribution of the radium may produce considerable discrepancies in the expected tissue dose as calculated for an assumed uniform packing. It would seem important, therefore, for those employing radium, first to study their radium supply to assure themselves that their sources are properly packed and of the strength and filtration designated by the manufacturer.

It is the purpose of this paper to describe a simple method of making autoradiographs of radium needles and tubes which can be carried out by anyone acquainted with the fundamentals of radiography and to present illustrations of defects and inequalities of radium distribution, which seem to emphasize the importance of such a study.

Certificates regarding the purity and quantity of the radium are usually supplied by the manufacturer or can be obtained from the U. S. Bureau of Standards. Such certificates are often taken as assurance that all sources are of the stated strength and filtration. However, in order to decrease the cost of measuring the radium content of sources and reduce exposure hazards in handling, it has been the practice of the U. S. Bureau of Standards to measure several radium sources of similar active length and content as a group rather than individually. For instance, ten radium needles of a stated strength of 2 mg. each and 3 cm. active length may be measured together, and a certificate supplied stating that the total radium content of the ten needles is 20 mg. Such a certificate does not necessarily indicate that each of the needles is 2 mg. strength, or that the radium is uniformly packed along its active length.

This additional information can be obtained by making autoradiographs of each radium needle and tube. Technics have been described by Griffith (2) and by Russ and Clephan (6) for making autoradiographs, and Clephan (1) has published a suitable formula which can be used as a guide for determining the exposure time required for needles and tubes of varying strengths, diameter, active length, and filtration, with a constant multiplication factor based on the speed of the film emulsion used.

This formula is as follows:

$$T = \frac{10(1 + S)^2}{M} \times L\sqrt{D}$$

where

M = radium content in milligrams
L = active length in centimeters
S = screenage or wall thickness in millimeters of platinum
D = diameter of bore in millimeters
T = calculated time of exposure in seconds.

The factor 10 represents the constant

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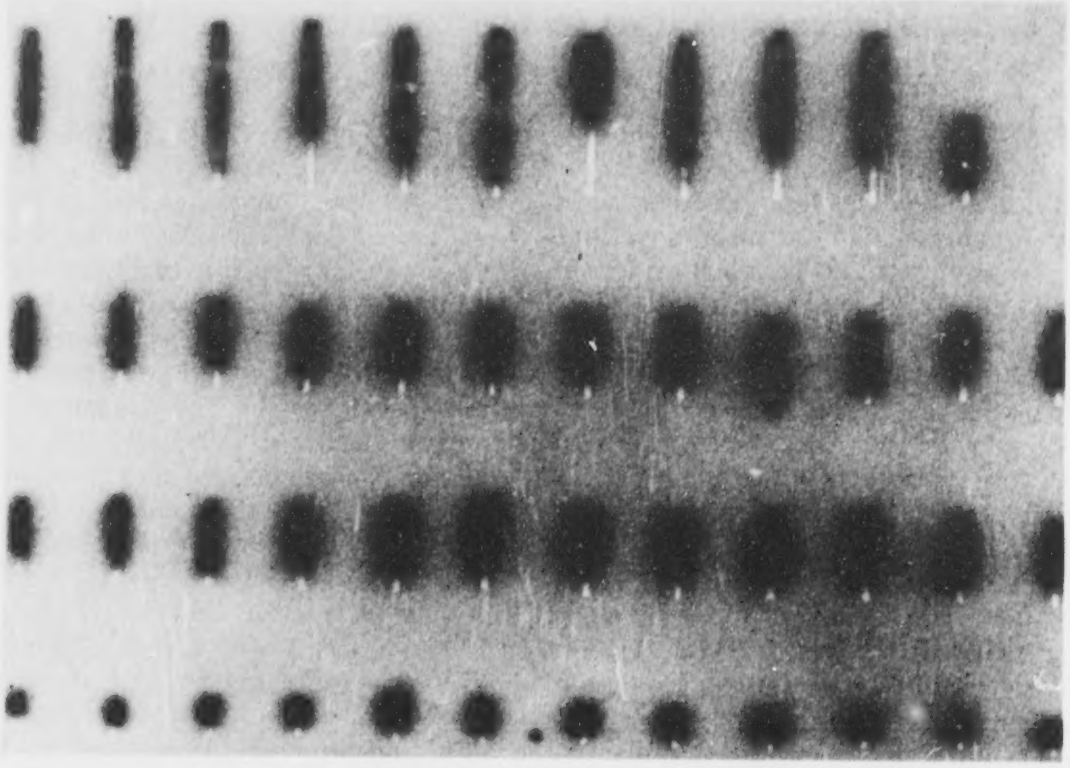

Fig. 1. Autoradiograph of a supply of poorly packed radium needles showing rather marked inequalities in radium distribution.

suitable with the use of Ilflex films, whose emulsion speed appears to be similar to that of Eastman non-screen film.

The technic which we have found to produce satisfactory autoradiographs is simple and easy to carry out and requires no material other than that available in any department of radiology. The radium holder is made by cutting slots of the size of the radium needles or tubes in a piece of cardboard obtained from a film package. Two strips of ordinary Scotch tape are pasted over the underside of these slots to prevent the radium sources from dropping out of their respective slots when the holder is moved. The adhesive base also prevents the sources from rolling about if the slots are slightly large in size.

Working behind a lead protective shield, the radium sources are placed in the various cardboard slots. This radium holder is then transported to one of the radiographic rooms on a long-handled carrier or cart, advantage being taken of distance, wherever possible, to keep the exposure of the individual handling the radium within the recommended permissible dose level of 6.25 milliroentgens per hour. Preliminary dosage calculations are made by the inverse-square law for the particular quantity of radium being handled, so that the dose level at the working distance is known at all times.

A film wrapped in black paper is centered beneath the x-ray tube on the radiographic table. The radium holder is then brought into the radiographic room, which is darkened except for suitable safe lights. The black paper is removed from the film and the holder placed directly in contact with the film. Eastman Kodak screen film was used in our studies and the formula as published by Clephan was utilized for determining a suitable exposure time, assuming a constant multiplication factor of 20.

After the desired exposure has been made, the radium holder is returned to the radium room and the film is developed for five minutes at 68° F.

Any needles or tubes which appear to be defective or show a marked inequality of

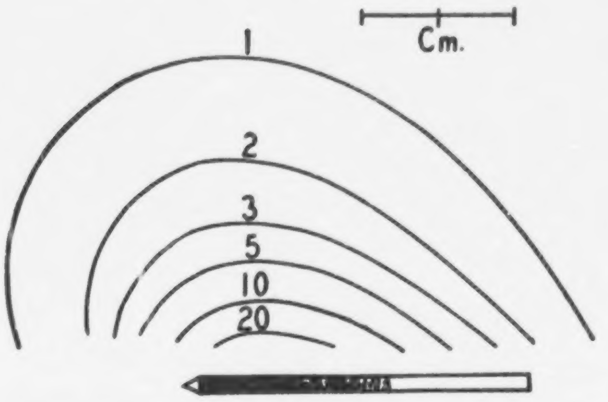

Fig. 2. Distortion of the isodose curves, in gamma roentgens per hour, about a 2-mg. needle, resulting from a faulty radium distribution.

radium distribution are identified by their respective positions on the autoradiograph and radium holder. These are removed and tagged as unsatisfactory for clinical use before being placed back in the radium safe. They are later returned to one of the radium companies for reconversion.

An autoradiograph of a supply of needles showing rather marked inequalities of distribution is shown in Figure 1. There is an undesirable variation in the strength of needles of similar active length and in the uniformity of the distribution along this active length. Leakage can be seen occurring from the broken end of one of these needles.

Variations in the uniformity of the radium packing causes a marked distortion of the radiation distribution in relation to the needle, as shown by the isodose curves about one of these needles (Fig. 2). These needles were prepared many years ago, before suitable packing technics had been devised.

The more modern methods of loading, particularly the cell method, tend to reduce the chance for errors in packing. Autoradiographs of some suitable cell-filled and direct-filled needles and tubes are shown in Figure 3.

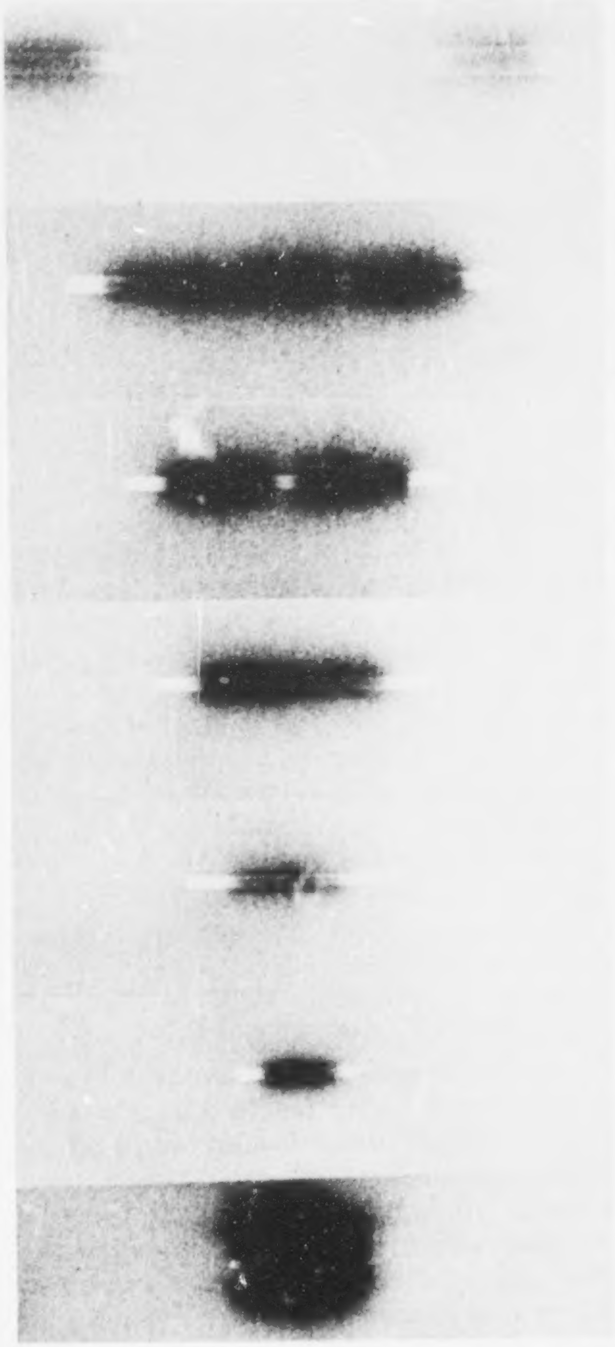

Fig. 3. Autoradiograph of cell-filled and direct-filled needles and tubes with satisfactory radium distribution.

The high intensity about the lowermost needle is largely due to unfiltered beta radiation passing through the 0.2-mm. platinum filter. This type of low-filtered, high-intensity needle was used years ago in interstitial therapy, but it was soon found that the high dose along the needle track led to excessive necrosis in the immediately adjacent tissue and prevented the delivery of adequate radiation to more remote tissues. This practice has now been almost universally abandoned in favor of low-intensity needles with a linear strength of not over 1 mg. per centimeter of active length, and a wall thickness of 0.5 mm.

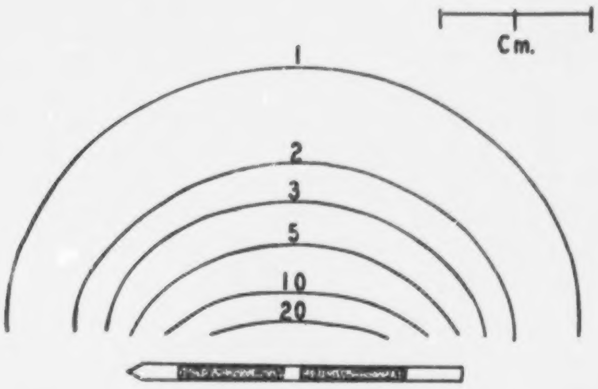

Fig. 4. Isodose curves in gamma roentgens per hour about a 2-mg. cell-filled needle of 3 cm. active length, with satisfactory radium distribution.

platinum. This filtration is sufficient to remove all of the beta radiation.

Higher filtrations have been advocated, but they tend to increase unduly the bulk of the needle, and the additional improvement in distribution and quality of the radiation does not appear to influence the biological effect.

The isodose curves about two low-intensity cell-filled radium needles, with uniform packing, are shown in Figures 4 and 5. Needles of this type give the most satisfactory results in interstitial therapy.

SUMMARY AND CONCLUSIONS

Autoradiography is an important and useful method of testing radium preparations for defects and uniformity of packing. Certificates, as issued by the manufacturer or by the U. S. Bureau of Standards, serve as a guarantee of the purity and quantity of radium, but do not give the purchaser any assurance regarding the uniformity of packing, and in some cases do not necessarily indicate the strength of the individual sources. This additional information can be obtained through autoradiographs.

A simple method of making autoradiographs of radium sources is described, with illustrations of poor radium distribution and satisfactory radium distribution in direct or cell-filled needles. Diagrams of the isodose pattern about a needle with poor radium distribution and two needles with uniform distribution are included to demonstrate further the importance of proper radium packing.

Physicians using or supervising the use of radium in radiation therapy should study their radium supply at intervals of five to ten years to reassure themselves that their radium sources are free of defects, uniformly packed, and of the stated strength and filtration designated by the manufacturer. Purchasers of radium in the future should insist upon a guarantee relating to distribution of the radium in the needles and tubes, as well as upon certificates of purity and quantity.

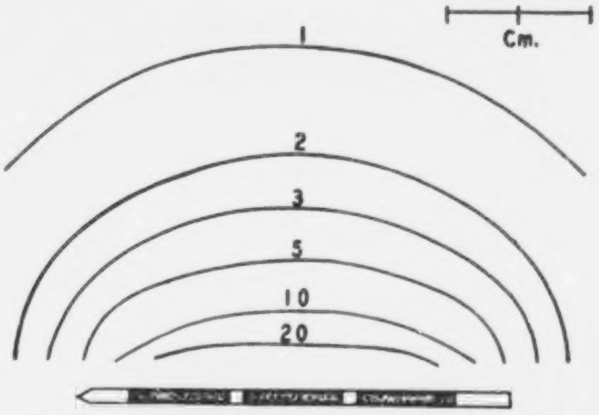

Fig. 5. Isodose curves in gamma roentgens per hour about a 3-mg. cell-filled needle of 4.5 cm. active length, with satisfactory radium distribution.

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TESTING RADIUM CAPSULES FOR RADON LEAKAGE

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RADIUM continues to be used in many hospitals, principally for intracavitary and interstitial γ-ray therapy. The use of encapsulated radium for external γ-ray therapy has been largely supplanted by high curiage Co^{60} teletherapy units, high voltage roentgen-ray machines and particle accelerators arranged to produce roentgen rays.

A limited industrial use continues but the total quantity involved, compared with the quantity used for medical purposes, is relatively small. Because of the 1,620 year half-life of radium, it is very convenient as a calibration source for radiation survey instruments and a number of organizations possess radium for this purpose. As mentioned above, it is used to a limited extent for industrial radiography, even though the reactor produced radioisotopes, Co^{60} and Ir^{192} are much less expensive.

The highly expensive decontamination procedures required in a hospital radiology department when a radium capsule ruptured has been described previously by one of the present authors and a colleague.[4] Potential hazards from spills are shared by our hospitals with industry; the most expensive spill on record resulted when an air-lifted sealed radium calibration source ruptured in a factory. The total cost of this accident was approximately $250,000 at the time settlement was made.[4] It may have increased since then because of many years of vacancy of the building involved.

Careful handling, periodic examination, periodic re-encapsulation and leak testing to check the integrity of the hermetic seal of the capsules are measures that should be taken by all users of "encapsulated radium sources" to prevent contamination with radon daughters, to reduce the chance of an accidental spill of the salt and to ensure the expected γ dose per milligram of radium content.

It is the purpose of the authors to describe a more sensitive leak test method for hermetically sealed radium cells, tubes and needles so "leakers" may be found and taken out of service before any of the radium salt is spilled, and to eliminate the use of sources that are not delivering the expected γ dose at the earliest possible moment. Also given are the results of our leak testing over a period of several years.

CHEMICAL AND PHYSICAL CHARACTERISTICS OF RADIUM

The physical and chemical characteristics of radium and its decay products have been extensively studied since the Curies separated and named radium in 1898. This information is given in detail by several authors[1,3,10] and only those properties having direct bearing on the problem of radium spills and leaking containers will be treated here.

1. The radium salts ($RaBr_2$, $RaCl_2$, or $RaSO_4$) are in the form of a finely divided, dry powder when loaded into the tubes or needles.

2. In its radioactive disintegration radium changes into radon, a "noble" gas, by emission of α particle.

3. Radon in turn, transforms through a series of 7 radioactive decay products, headed by radium A (polonium 218), ultimately becoming a stable lead 206. Each of these decay products of radon is a radioactive isotope of either thallium, lead, bismuth, or polonium, and hence is a solid at room temperatures.

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4. Radium and radon are α emitters with no important γ radiation. The γ rays associated with radium sources are in fact emitted principally in the β decay of the daughter products radium B (lead 214) and radium C (bismuth 214). Therefore radium and radon must be in equilibrium with their γ-emitting daughters to be useful as a source of γ radiation.

When a radium tube or needle loses its hermetic seal, it continuously leaks radon gas which, if at all confined, will remain long enough to undergo radioactive decay and produce particulate radon daughters that will adhere to the sides of the lead storage container, the transport container, and will, ultimately, contaminate the "L" block, applicators and other equipment. Escape of radon gas from a "sealed" radium container should act as an immediate red flag, particularly if the capsule is closed with a press-fit plug that has been soldered to effect a gas tight seal. Escaping radon indicates that the solder has deteriorated and the plug may drop out, as has been the case in some very serious spills reported in recent years.

When a radium spill occurs, the physical state of the encapsulated radium salt—as an extremely fine dry powder—and the high radiotoxicity of this radioisotope combine to accentuate the seriousness of the exposure to individuals in the vicinity, to inhalation of the finely divided dust, as well as the contamination problem. Unless prompt and stringent control measures are taken, past experience has shown widespread tracking almost invariably occurs with consequent high decontamination expense.

TYPES OF RADIUM CONTAINERS

American style radium containers are commercially available as 90 per cent platinum, 10 per cent iridium alloy cells, needles and tubes, in a wide range of sizes and radium content, best suited to the varying needs of the radiologist. The extremely small, easy to lose, platinum-iridium cells are now used widely by hospitals. The smallest have an outside diameter of 1.0 mm. with an external length of 11.5 mm. Cells have been available from some suppliers with wall thicknesses of either 0.2 mm. or 0.5 mm., with the radium content ranging up as high as 10 mg., the diameter up to 1.90 mm. and the length up to 64 mm.

Inasmuch as 0.5 mm. of gold or platinum is needed to filter out most of the primary β particles from radium in equilibrium with its decay products, most radium dosage charts are calculated on the basis of 0.5 mm. platinum equivalent filtration; the 0.2 mm. wall thickness cells are normally loaded into hollow needles or tubes for use, having 0.3 mm. Pt-Ir alloy wall thicknesses to give a total filtration of 0.5 mm.

Containers for larger quantities of radium, in the 25 to 100 mg. range, have 1.0 mm. thick walls, are considerably sturdier as a consequence and much less likely to be bent or crushed.

Some needles and tubes were direct-filled and sealed; others were loaded with sealed cells and then the outside needle or tube was again sealed, giving an additional factor of safety. All preparations from one supplier in the form of needles and tubes that have been shipped during the past several years on a direct sale basis have been sealed with gold solder into cells which were in turn gold-solder sealed into an outer container in the form of a needle or tube.[6] This supplier furnishes radium sources for industrial radiography in all monel cylinders that are usually enclosed in duraluminum cartridges when in use.

Surface applicators (plaques) are available in almost any size and strength desired by the radiologist. They are designed to contain a given size and number of cells, or needles, so the hermetically sealed unit is still the individual cell or needle. Monel metal nasopharyngeal applicators are widely used. Though apparently very sturdy containers, we have found some which were leaking radon.

Occasionally, a user of radium is found with the "European" style of encapsulation. One type is the sealed quartz capsule placed inside a metal container with a screw thread cap. Experience has shown that this form of encapsulation can be hazardous. One such source was involved in a very expensive spill.

Radium for industrial radiography marketed by another supplier was encapsulated in silver, right cylinders, in the form of the sulfate, with all joints sealed by brazing. Usually, the sources were placed in a steel outer container if a magnetic handling pole was to be used, and in aluminum "plumb bob" type containers if they were to be handled by string.

NEED FOR LEAK TESTING

The average Community Hospital has about 100 mg. of radium divided into from 15 to 20 individual cells, capsules or needles, if it has any radium at all. The larger hospitals associated with Medical Schools, or specializing in radium therapy, may have a much greater inventory—400 to 500 mg. divided into 100 or more containers. Only one radon gas "leaker" in the lot can spread low-level radon daughter contamination widely. Surveys have shown that the interior surfaces of a hospital's lead storage container, its carrying container, applicators, and usually much of the handling equipment will be contaminated to a low level though no more than 1 or 2 capsules were found to be leaking. This contamination may not be serious as a health hazard—the amount of long-lived radon daughters that is likely to be inhaled or ingested is extremely small. Nevertheless, it is an undesirable situation; the leaking container is not producing the γ dose on which the radiologist is basing his calculations, and the potential hazard of a spill of the radium salt is increased greatly, when the capsule is defective to the extent that it is leaking radon.

TYPES OF LEAK TEST

Leak testing of radium capsules and needles was accomplished, originally, by laying them on cotton for at least 24 hours and then measuring the amount of activity on the cotton with the best instrument at hand. A refinement on this method was to seal the radium capsules individually in small test tubes with a wad of cotton for a suitable period. To overcome the large self-absorption of the cotton when counting α activity, the cotton was replaced with filter paper disks placed at the top of each test tube so as to avoid physical contact with the capsule or needle. (Experience has shown that α counting is much more sensitive then measuring the β and γ radiation.) The filter papers then were counted for α activity from the radon daughters absorbed onto the surfaces.[4] From 24 to 72 hours was found to be a sufficient length of time to keep the filter papers sealed in the test tubes with the capsules. A portable Samson or Gas Flow Alpha Survey Meter was used to measure α activity absorbed onto the filter papers.

The above described methods have certain disadvantages. Because of lack of sensitivity, frequently, needles or cells leaking radon only slightly are difficult to detect with any certainty. Also, it may be inconvenient to wait 24 hours, or even over night, to check the filter papers for activity when making field surveys. An electrostatic method of testing for radon leakage described below satisfies the requirements for greater sensitivity and eliminates the 24 hour wait before counting the filter papers.

The electrostatic leak test apparatus shown in Figure 1 was built by one of the authors at the M.I.T. Radioactivity Center, to leak test low level radium containing mixtures sealed in small vials. The 1 inch inside diameter, 6 inch long glass tubes are sealed when in use with rubber stoppers dipped in Dekadhese to prevent absorption of radon. The thin copper sheet electrodes mounted on each of the rubber stoppers collect a large fraction of the first daughter products of all radon atoms which decay in the free volume of the glass tube, because these daughter products carry a net positive charge immediately after their formation by α decay. The 300 volt battery which provides the electrostatic collecting potential is a type readily available, and its life when used for this purpose is limited only by the shelf life. Any other direct current supply of the order of 300 volts is, of course, equivalent. Electro-deposition of radon daughters occurs on both electrodes; however, repeated counts have shown the amount of α and β activity on the cathode (negative electrode) to be about 10 times greater than on the anode. This is to be expected because of the net positive charge of freshly formed daughter products. Therefore, in routine use only the activity on the cathode is counted. With this device a leak corresponding to the radon produced by 0.0004 mg. of radium can be easily detected by counting the radium B and radium C β-ray activity of the negative electrode, with an end-window Geiger-Müller counter, after 1 hour of storage of the radium source in the collecting tube. For short collection times the growth of radon in the collector from a leaking radium source is linear with time. Hence, a 1 per cent leak in a 1 mg. radium needle is easily detected with this device with only a 3 minute collection time. The β activity of the collecting electrode, after removal from the collector, decays with time in accord with the Bateman equations[2] controlled mainly by the 27 minute half-period of radium B. If radium, rather than merely radon, has leaked out of the vial or needle being tested, the collector will continue to accumulate daughter products after removal of the vial or needle.

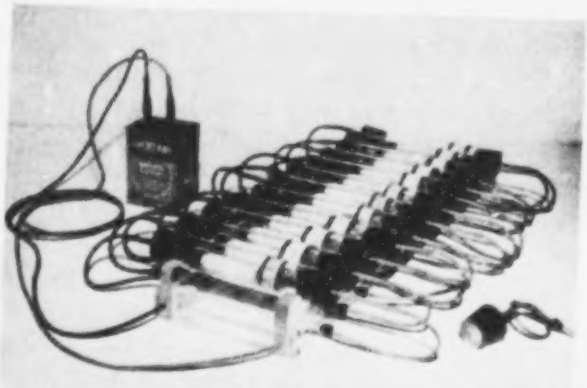

Fig. 1. Electrostatic leak test apparatus used at M.I.T. Radioactivity Center to test for radon leakage from low level, radium containing mixtures sealed in small vials, as developed by R. D. Evans as a part of Manhattan District and Atomic Energy Commission contract No. W-22-075-eng-13.

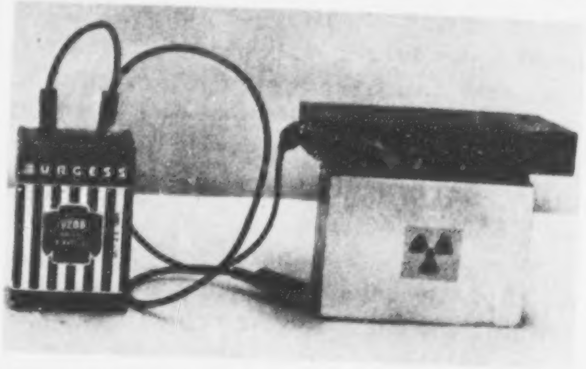

Fig. 2. Modification of electrostatic leak test apparatus, capable of leak testing 30 therapeutic radium sources at a time. Shielding not shown.

Figure 2 shows a modification of the leak test apparatus that has been found to be more suitable for field use to test encapsulated radium in hospitals. Thirty ¼ inch (7 mm.) inside diameter polyethylene tubes 2¾ inches (7 cm.) long were cut from stock and brass plugs to serve as electrodes (cathodes) were provided with necks for easy gripping with 6 inch long lock forceps when inserting or removing them when radium is present in the tubes. During leak tests the radium containing tubes are placed in the small rectangular wooden block, resting on coiled spring contacts soldered to brass bus bars. By placing a 1 inch thick lead brick on the cathodes, good electrical contact is assured for top and bottom electrodes in each tube and some shielding is provided. The battery is connected with leads having an alligator clip on one end and a banana plug on the other. The 1 inch thick lead brick serves as the negative electrode.

In use, lead bricks and the hospital's "L" block are utilized to reduce radiation dose rates to safe levels outside the radium storage room. If no lead bricks are available, the lead-lined deep roentgen ray therapy room may be used, usually, late in the day.

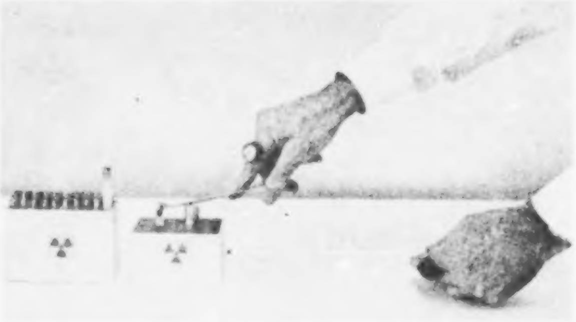

Fig. 3. Split lead block vise used to hold polyethylene tubes during removal of cathodes. Also shown are polyethylene tubes and wooden block used to hold them during leak testing.

The capsules must be held while inserting or removing the brass plugs (cathodes) because of the snug fit, as shown in Figure 3. To reduce the radiation exposure to a minimum while doing this, a small vise was devised, consisting of a split lead block with the two pieces grooved to hold the tubes. The larger of the two pieces is toward the handle used to clamp them together. The pedestal was added only recently to serve as a fulcrum when withdrawing the cathode plugs from the polyethylene tubes. It was found to appreciably reduce handling time and hand exposure by facilitating cathode removal. Repeated checks with quartz fiber dosimeters and wrist film badges have shown the hand dose to be well below the maximum permissible dose given as a guide by N.C.R.P. when the lead block vise and lead bricks are used around the block containing the polyethylene tubes, with no more than two surveys per week.

Though much shorter collection times are satisfactory, as mentioned above, it has been our practice to leave the radium sources sealed in the tubes for at least 4 hours to reduce the chance of missing a source with only a slight leak. The collecting surface of each cathode plug is checked for presence of radioactivity with either a Gas Flow Alpha Meter or a Samson Alpha Meter. The Samson meter is preferable because it "sees" β particles also, and permits a lower threshold of detection when sources leaking only slightly are encountered.

LEAK TEST RESULTS

Data from radiological safety surveys in 24 hospitals and 3 doctors' private offices showed 37 sources (radium capsules, cells and needles), out of a total of 664 leak tested, to be leaking radon gas. In at least 2 instances, one or more of the radium capsules were permitting escape of the parent radium. The total quantity of radium contained in the 664 sources was 4,318 mc.

Almost without exception, at least some of the applicators, all of the storage containers and much of the handling equipment were found to be contaminated where leaking capsules or needles were detected. In 5 of the hospitals surveyed, the area in which the radium was stored was contaminated. In general, the area of contamination has been found around the "L" block, but in one case it was spread over 300 square feet of floor surface. It is the usual thing to find responsible hospital personnel unaware of the defective radium sources. It is hoped that this will improve as more and more hospitals obtain portable radiation survey instruments that are suitable for use in checking for presence of unwanted radioactivity in carrying containers, applicators, vises and similar handling equipment. Efforts at decontaminating such equipment as carrying containers, applicators, and handling forceps that were brought back to the laboratory for experimentation have shown that in the usual case it is less expensive to discard this material as radioactive waste and buy new replacements.

When leak testing of hospital radium was first made a part of our radiological safety surveys several years ago, some inventories of radium were found to be in extremely poor condition. For example, one survey showed 6 out of 8 capsules possessed by the hospital (100 mc of radium) to be leaking. These capsules were 23 years old and had never been leak tested before. In another hospital, 7 out of 29 radium sources were found to be leaking. The exact age of these 29 needles and capsules could not be determined, but it was about 8 years. Fortunately, the situation has improved rapidly in the past 5 years in the institutions with which we are familiar. Many inventories of hospital owned, or rented, radium tubes and cells have been leak tested recently, with no "leakers" found.

On several occasions leaking radium cells and needles have been brought into our laboratory, after detection in routine surveys, to see if the cause of leakage could be ascertained. Usually, no defects could be seen with the naked eye. However, the cause of failure of the encapsulation was visible with the aid of a microscope in 2 cases. One leaking cell, originally containing 1.33 mg. of Ra, had two "pits" in the end plug, possibly caused by corrosion or escape of gas at the time of sealing when too much heat was used. This cell was leaking radon freely; evidently the "pits" penetrated into the interior chamber. Figure 4 is a photomicrograph of a 5 mg. radium needle with a loose plug. The hazard involved in handling a needle in this condition is obvious. If the plug dropped out during handling, widespread contamination could easily result. This needle was leaking radon freely. Therefore little, if any, gas pressure within the interior chamber existed to force the plug out.

FREQUENCY OF LEAK TESTING

It has been our practice for a number of years to recommend annual leak testing of all sealed radium sources. Perhaps more frequent leak testing would be desirable but consideration must be given to the radiation dose received in handling the pieces, especially when a large percentage of the supply is loaded in Heyman applicators that are badly corroded and the screw caps are difficult to remove. If sufficient manpower is available so as to spread the exposure among them, leak testing every 6 months probably should be done on frequently used radium. One large hospital has its entire radium inventory (over 490 mg.) leak tested every 6 months. The task is assigned to either a resident radiologist or medical physicist and it is considered a part of his training.

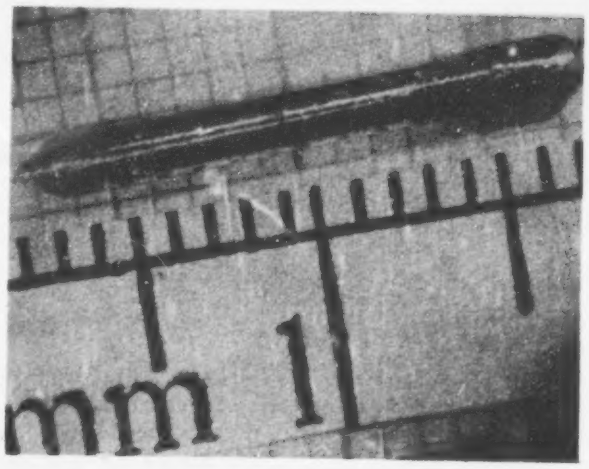

Fig. 4. Photomicrograph of leaking 5 mg. radium needle with loose plug. Note separation of plug from body of needle at upper right.

The 6 months' schedule for leak testing mentioned above is in marked contrast to the situation usually found, where no schedule at all exists for routine radium leak testing. For example, 4 hospitals surveyed in recent months possessed a combined total of over 1,000 mg. of radium, divided into about 200 individual cells, needles and tubes. None of the radium was leak tested on a routine basis. It is of some interest that 3 out of the 4 hospitals had leaking tubes or cells in their radium inventory. Two leaking cells were found in one of the hospitals that had sent all its radium back to the company from which it was purchased, for leak testing and re-calibration within the year.

PERIODIC RE-ENCAPSULATION

It is not our purpose to treat in detail the problem of gas pressure in sealed radium containers. Nevertheless, our experience indicates that no opportunity should be overlooked to impress upon the minds of all users of sealed radium containers the necessity for relieving the built up gas pressure, periodically. J. E. Morgan[8] has recently published an excellent discussion of the hazard of explosive rupture caused by inevitable accumulations of helium. His calculations showed that a pressure build-up of helium of 1.09 atmospheres per year will occur in typical medical radium needles, cells and tubes. His conclusion regarding frequency of re-encapsulation was not specific; periodic re-encapsulation at intervals of a few years was recommended. An arbitrarily selected interval of 10 years has been recommended by us since the start of our leak testing. This is considered a conservative time limit for re-encapsulation in sealed radium sources made of Pt-Ir alloy.

SUMMARY

Sealed radium sources are used, predominantly, for medical purposes in this country. Expensive spills of the radium salt are on record. Careful handling, periodic examination, testing for radon leakage and re-encapsulation should be provided for all sealed radium sources. Aside from the potentially severe health hazard, careful attention to maintenance of the hermetic seal of radium sources is important for 2 reasons: (1) a spill of the finely divided radium salt can be highly expensive to clean up and (2) the source does not deliver the gamma dose expected of it unless the radon daughters are in equilibrium with the parent radium.

A new, more sensitive, electrostatic precipitation method of leak testing sealed radium sources has been developed and field tested for several years. This method is far more sensitive than earlier methods and a time-saver during field surveys.

Radiological safety surveys in 24 hospitals and 3 doctors' offices revealed 37 leaking radium sources out of a total of 664 leak tested (total of 4,318 mc). Leaking sources contaminate storage safes, handling equipment and applicators.

It is the conclusion of the authors that sealed radium sources should be tested for radon leakage annually. We also feel that such sources should be routinely re-encapsulated every 10 years to relieve the internal gas pressure caused by inevitable accumulation of helium when α particles capture electrons and become helium atoms.

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Radon Film Badge

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Introduction

THE DETECTION and quantitative measurement of radioactive radon gas is important in enclosed spaces, where concentrations in air can reach levels in excess of $(\mathrm{MPC})_a$, and for testing sealed radium sources for leakage. The ability to measure low concentrations of radon gas without expensive equipment or special training may also have industrial applications, e.g. in oil exploration or uranium prospecting.

Radon, an inert gas that readily diffuses through earth, air and crevices, decays by the emission of an alpha particle. After the emission of an alpha particle, the atom exists for a short period of time as an isotope of polonium which subsequently decays by alpha emission to other "daughter products" of radon. The principle of detection described herein is based on the use of film to measure alpha particles from radon gas and its daughter products.

Physical description

The radon film badge (Fig. 1) consists of a small ($1\frac{7}{8}$ in. × $1\frac{1}{4}$ in. × $\frac{17}{64}$ in.) light-tight holder with small bores extending outward from a central recess in a plurality of directions to the exterior of the holder. A sheet of alpha sensitive film is positioned inside the badge and is separated from the central recess by a light-tight window sufficiently thin to permit substantially unimpeded passage of alpha particles. The radon gas diffuses into the central recess where the alpha particles from radon and its daughter products pass through the window and into the film emulsion. The number of alpha particles is determined by counting the alpha tracks in the processed film emulsion. The number of tracks is directly proportional to radon gas exposure.

Test and evaluation

Calibration of radon concentrations

Preliminary screening of prototype film holders was conducted in the laboratory using radon gas generated by bubbling air through a solution containing a known amount of radium. Final prototype selection, evaluation and calibration were accomplished in a uranium mine in the Grants, New Mexico area.

Gas concentrations were measured by collecting air samples in evacuated flasks which were subsequently analyzed for radon gas by the Massachusetts Institute of Technology. The radon daughter products were collected on a filter paper from a known volume of air and the ionization measured after a limited period of decay. This latter technique is the one most commonly used in the mining industry to measure a "working level", which is equivalent to 0.1 pCi of $^{222}\mathrm{Rn/cc}$, assuming the radon and its daughter products are at equilibrium.

The response of the radon badge was related to radon gas concentration and to the concentration of radon daughter products attached to dust particles. Response factors were calculated by dividing the number of alpha tracks observed per 7 mm² with a binocular microscope by the number of hours exposure at $(\mathrm{MPC})_a$ based on the measurement of radon gas, R_G, and radon daughters, R_D. For these calculations, $(\mathrm{MPC})_a$ was assumed to be 0.1 pCi of radon per cc of air and the daughter products associated with this amount of radon at equilibrium. In actual practice the daughter product concentration is seldom at equilibrium with radon gas; therefore, R_D is higher than R_G. The ratio $(R_G/R_D)(100)$ is the per cent of equilibrium.

Response–gas vs. daughters

To separate the response of the badge to radon gas from its response to radon daughters, badges were exposed to a fairly stable concentration of radon and radon daughters flowing at a rate of 60 ft/min for a total time of 21.5 hr. The ratio of radon daughters to radon gas was 75 per cent of equilibrium during this test. Badges were sealed after 0.25, 5.1 and 11.5 hr. Other badges were left unsealed for the entire exposure time. The "sealed" badges were considered sealed with respect to radon daughters but not necessarily sealed to radon gas.

From this test, the response of the badge to radon gas and its response to plate-out of radon daughters were calculated. The per cent of the total response due to plate-out was plotted vs. per cent of the total exposure time each badge was left open to daughter plate-out (Fig. 2).

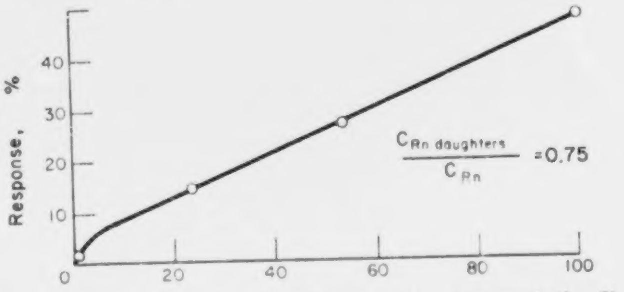

FIG. 2. Response due to plate-out of radon daughters.

The results indicate that for this set of conditions, when the air passages in the badge are open 100 per cent of the time, less than 50 per cent of the film response is due to plate-out of radon daughters other than the daughters produced by the decay of radon gas after it reaches the central cavity. This contribution from direct plate-out (vs. "in-growth") of radon daughters can vary from less than 10 per cent too over 50 per cent depending on environmental conditions.

Linearity of response

The number of alpha particle tracks observed in a 7 mm^2 area was plotted vs. concentration multiplied by length of exposure (Fig. 3). The response was linear with exposure. Thus, a single factor can be used to convert observed alpha tracks to total exposure. This factor for ^{222}Rn gas is 5.2 alpha tracks per pCi-hr/cc. The error terms shown (Fig. 3) are the standard deviations for each count.

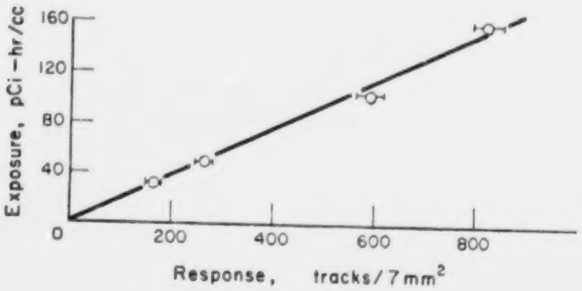

FIG. 3. Exposure vs. response.

Effect of badge orientation and air flow rate

The effect of air velocity on the radon response factor, R_G, was not significant in the range of 30–120 ft/min (Table 1). The response factor based on daughter products, R_D, was higher and more erratic.

Precision

To test precision, badges were exposed in duplicate to various total exposure levels. Data accumulated for varying exposure conditions indicated that R_G was reproducible whereas R_D was very erratic (Table 1). Most of the error associated with R_G in Table 1 was attributed to the variation in direct plate-out of radon daughters. However, this variation caused less than ±30 per cent over-all error even with an extreme set of exposure conditions.

Sensitivity

The sensitivity of the method is ultimately limited by the number of fields one is willing to look at under the microscope. The theoretical number of tracks that should be seen in a 7 mm^2 area of film after a 40-hr exposure at 0.1 pCi/cc has been calculated for ^{222}Rn. The recess in the center of the badge is 3.7 mm deep; therefore, the effective volume for a 7 mm^2 film area is:

$$(3.7\ \text{mm})(7\ \text{mm}^2) = 25.9\ \text{mm}^3 = 0.0259\ \text{cm}^3.$$

At a concentration of 0.1 pCi/cc, this volume contains:

$$(0.0259\ \text{cm}^3)(0.1\ \text{pCi/cm}^3)(2.22\ \text{dpm/pCi}) = 0.00575\ \text{dpm of}\ ^{222}\text{Rn}.$$

The alpha emitting daughters, ^{218}Po and ^{214}Po, establish equilibrium with the ^{222}Rn inside the badge and each contributes 0.00575 α/min for a total of 0.01725 α/min. However, only half these α-particles pass through the film, the remainder being emitted in the opposite direction. Thus a concentration of 0.1 pCi/cc should produce 0.0086 tracks/min, 0.52 tracks/hr and 21 tracks in 40 hr. The empirical response factor is 0.52 ± 0.14 tracks/hr, based on the data in Table 1.

The normal background of this film is approximately 4 tracks/7 mm^2. The actual background is measured for each film. $(\text{MPC})_a$ based on a 40-hr exposure can be detected with 99+ per cent confidence. Lower levels can be measured with longer exposure periods or by counting the tracks in a larger area of the film.

Image fading

The latent image fading in most applications is the same as for neutron film dosimetry.(1–3) Exposure periods up to one month have been used successfully under favorable environmental conditions. The effect of fading has been minimized in this laboratory by using optimum processing conditions.(3)

To evaluate use limitations under adverse conditions, badges were exposed in a uranium mine for 30, 72 and 168 hr. The badges were exposed in an area of high radon concentration, 95 per cent relative humidity, 62°F and an air velocity of 86 ft/min and then stored in another area in the mine where all the conditions were the same except that the radon concentration was very low. The control badge left in the low radon area indicated less than 5 per cent of the total exposure occurred in this area. The fading was not significant after 30 and 72 hr. After 168 hr in this environment, the films were damaged because the emulsion had stuck to the aluminized mylar window. For use periods longer than 72 hr in a humid mine environment, the badge should be redesigned so the film will not stick to the window.

Applications

The radon film badge is a sensitive detector for radon gas. The application of the device as a personnel dosimeter in the uranium mining industry is

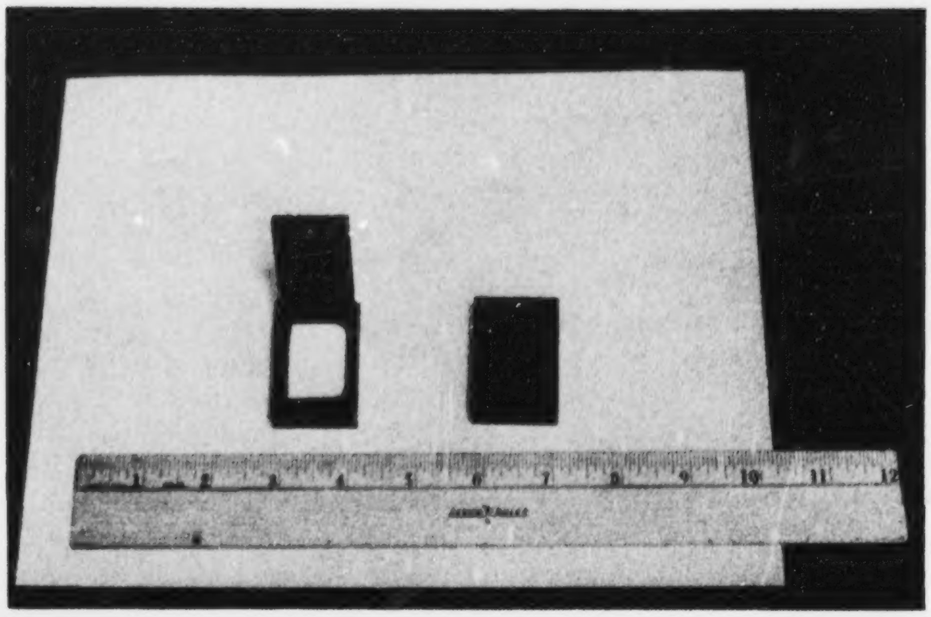

FIG. 1. Radon film badge

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immediately apparent but its use for leak testing of radium sources, for oil exploration and other less obvious applications may prove to be its greatest value.

Mining operations

For use in the underground mine, a weekly exchange frequency is recommended. A badge may be worn by an individual to document his total radon exposure. The badge may also be used as an area monitor to document average radon concentrations in various areas of the mine.

Table 1. *Effect of orientation and flow rate on response and precision*

Flow rate ft/min	Badge orientation*	Response R_D	Factors R_G
280	Normal	1.14	0.36
220	Normal	1.61	0.89
220	Normal	0.79	0.36
220	Normal	1.43	0.80
120	Parallel	2.38	0.48
110	Normal	0.97	0.51
110	Normal	0.68	0.44
110	Normal	0.97	0.51
90	Parallel	2.32	0.45
90	Normal	1.92	0.37
90	45°	2.06	0.40
60	Normal	0.73	0.52
60	Normal	0.69	0.50
60	Normal	0.76	0.60
60	Normal	0.61	0.48
60	Normal	0.79	0.59
60	Normal	0.84	0.63
60	Normal	0.82	0.58
60	Normal	0.69	0.49
Average		1.17	0.52
Standard deviation		0.60	0.14

* Orientation of small bores extending through the badge with respect to the direction of air flow.

Radium source leak testing

The procedure used to leak test a radium source must include a small container to trap the radon gas leaking from the source and shielding to reduce the γ-exposure to the badge. For example, a source stored in a small cylindrical lead shield can be leak tested as follows:

(a) Remove the top plug from the lead shield.

(b) Place the badge next to the lead shield and invert a small container, e.g. a coffee can, over the lead shield and badge. To obtain a better seal, the edges of the can should be smeared with surgical jelly, inverted over the source and shield on a smooth surface and then weighted on top so that the leaking radon gas is trapped under the can.

(c) After 4 hr, remove and return the radon film badge for processing.

Gamma exposure will not be erroneously interpreted as radon gas exposure but may make the alpha tracks difficult to count. Film have been read in this laboratory with up to 5 R of γ-exposure.

Radium–beryllium neutron sources may also be leak tested with the radon film badge. In this case, the badge or the source should be shielded so that the neutrons are moderated and absorbed for best results. The badge is designed so that proton recoil tracks produced by the neutrons can be distinguished from radon alpha tracks but a large number of neutrons does make interpretation more difficult.

Oil exploration

Since 1953 many articles have been published concerning radiation anomalies taking the form of a halo around producing oil and gas fields.(4–10) The oil has a blocking effect on the seepage of radon gas from the earth. Where there is no oil, the tortuous path of the gas through both pervious and impervious strata from source to surface is essentially vertical. Thus, the area above an oil body has a lower rate of radon gas seepage while the periphery of the oil field has a higher rate. By utilizing collection chambers in the suspected area to trap the gas seeping from the earth and using the radon badge to measure the concentration build-up in the chamber, it may be possible to define the boundary of an oil body more precisely than has been previously practical. With a simple but well designed collection chamber and a one-month exposure time, a radon concentration build-up of 0.01 pCi/cc can be measured with a ±30 per cent coefficient of variation. Higher levels than this have been observed to accumulate under buildings with a confined crawl space, thus the sensitivity should be adequate for this application.

Summary

A special film badge has been developed that is sensitive to radon gas. A sheet of alpha sensitive film is positioned inside the badge and is separated from a central recess by a light-tight window sufficiently thin to permit substantially unimpeded passage of α-particles. The radon gas diffuses into the central recess through small bores where it decays by α-emission. The number of α-particles is determined by counting the alpha tracks in the film emulsion. The number of tracks is directly proportional to radon gas exposure.

The precision of response for a wide range of exposure conditions showed a coefficient of variation of ±30 per cent. The sensitivity is adequate to detect (MPC)$_a$ ± 50% for ^{222}Rn (40-hr exposure) with 95 per cent confidence. The response is linear with exposure and is relatively independent of wind velocity. The badge may be used to leak test radium sources, monitor personnel for radon gas exposure, monitor areas for radon concentrations and for a variety of other applications.

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SOME DEFECTS IN RADIUM NEEDLES—THEIR DETECTION AND SOME CONSEQUENCES

By H. D. Griffith, B.A.
The Aberdeen Radium Centre

(*Received March* 20, 1936)

THE use of the autoradiogram, preferably with subsequent short exposure to visible light to give a shadow of the outline of the screen on the film, has become a recognised method of testing radium preparations for uniformity of packing.[1] In a discussion on the question of non-uniformities indicated by this method, doubt arose as to whether some of the observed effects could be ascribed to small variation in thickness of the walls of the platinum screens. Since most of the blackening of the film in contact with a radium needle is due to radiations of very mixed hardness, it is hardly possible to calculate how much effect a given variation of wall thickness might be expected to produce on a film in contact with the metal; direct experiment shows that this criticism of autoradiograms as tests of uniform packing is without foundation. Autoradiograms (Figs. 1 and 2) were obtained of two radon preparations, in thin glass capillary tubes as used for making seeds, laid on sheets of platinum foil below which were pieces of X-ray film. In Fig. 1, the capillary containing the radon was uniform, but was placed over a layer of platinum foil 0·30 mm. thick, half of which was covered with a second layer of platinum foil 0·06 mm. thick. The effective screenage under the right half of the radon source was therefore 0·36 mm., while under the left half it was 0·30 mm. No density-step can be seen in the autoradiogram to indicate this considerable variation of screenage. In Fig. 2, the screenage was provided by a layer of platinum foil of uniform thickness, 0·30 mm.; the source consisted of two radon capillaries end to end, one, R, having linear activity 38·8 mc. per cm., the other, R_2, 10·4 mc. per cm. The variation of linear activity is clearly shown by the autoradiograms.

Detailed investigation was made of the needle shown in Fig. 3, a 2 mgm. needle, 44 mm. in over-all length, in 0·8 mm. platinum screen. The measured total activity corresponded to 1·98 mgm., but the autoradiogram indicated that the bulk of the radium lay in the rear cell; the total charge was correct, but it was badly distributed. By adjusting the time of exposure of the film by trial until the photographic densities agreed with those produced by a 10-minute exposure to a well packed needle, and assuming a simple reciprocity law, the linear activity in the rear half of the needle was estimated as 1·2 mgm. per cm., or 1·5 mgm. in the 13 mm. length of the cell; the remaining 0·5 mgm. would appear to be concentrated in about 6 mm. near the middle of the needle.

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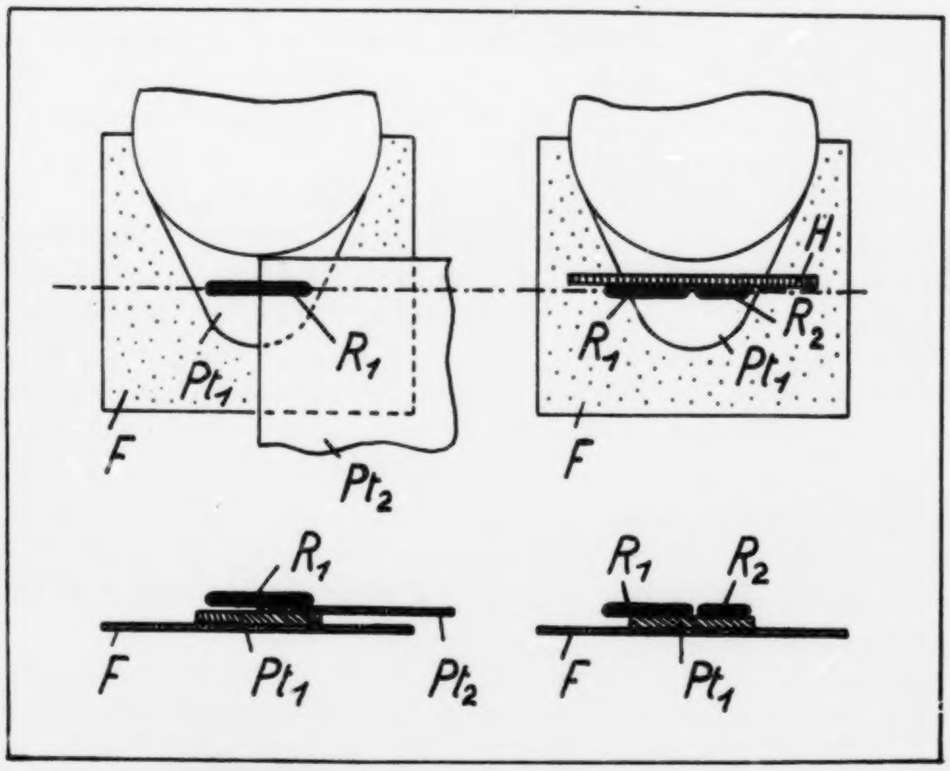
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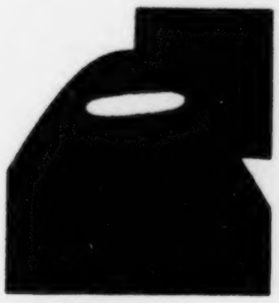


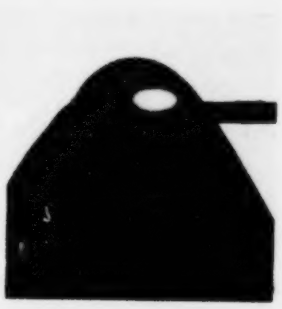
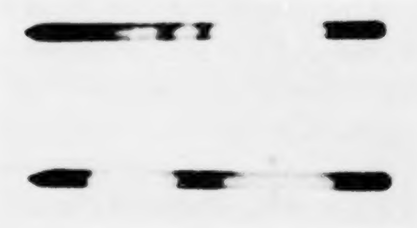


Fig. 1.
Effect of non-uniform screenage on autoradiogram.

Fig. 2.
Effect of non-uniform linear activity on autoradiogram.



Figs. 3 and 4. Badly packed and satisfactorily packed needles.

Fig. 4 shows a properly packed needle of the same dimensions. It should be noted that these figures indicate a linear activity in the rear of the defective needle of double that found in the standard needle, which has only 0·625 mgm. per cm.

The isodose distribution for the defective needle was determined with an ionisation chamber of volume 40 cu. mm., the walls being of graphite 4 mm. thick, enabling the chamber to be brought to a distance of 7 mm. from the axis of the needle. The indicating system was a thread electrometer arranged as described by Goldhaber and Griffith.[2] Fig. 5 gives the intensity distribution along a line parallel to the axis, at 1 cm. distance, and also a similar measured curve for a properly packed needle. In order to see whether the estimate of the linear intensities obtained from the autoradiogram could be confirmed, the intensity distribution curve for a pair of sources in line, for points 1 cm. from the axis, was calculated from Sievert's tables,[3] the linear activities being as indicated from the autoradiograms. The resulting curve, shown in Fig. 6, is seen to follow the same course as the observed curve. It would appear, therefore, that rough estimates of the degree of non-uniformity of packing can be made from autoradiograms by the technique suggested. Fig. 7 shows a faulty

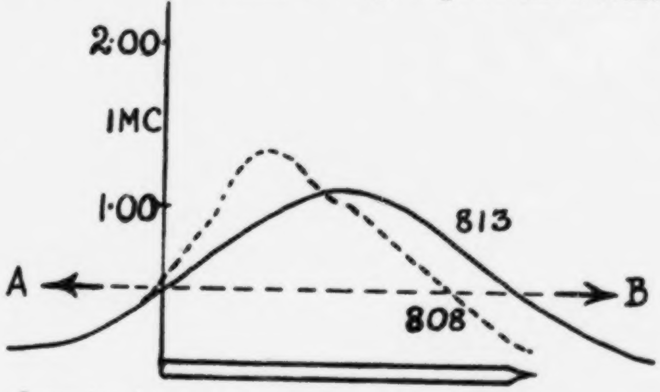


Fig. 5. Intensity distribution near well packed and badly packed needles.

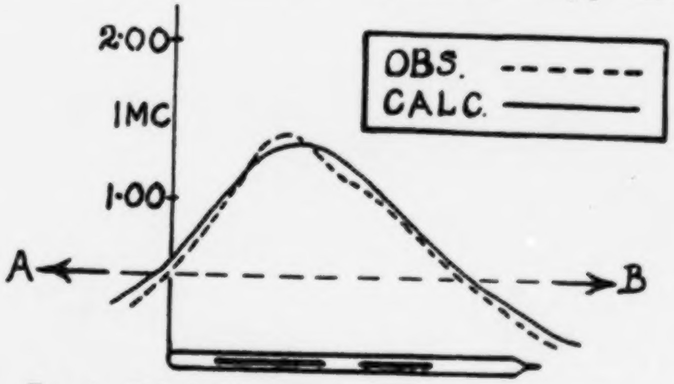


Fig. 6. Calculated and observed intensity distributions.

intra-uterine applicator and its isodose curves in a plane through the line AB. 1 cm. from the inside of the bend.

It is interesting to note that leaks in radium tubes can sometimes be demonstrated on the autoradiograms. A crevice from which radon has slowly diffused will be coated with radium A, the alpha particles from which will give a characteristic, sharp-edged fan of dense blackening. Two cases illustrating appearances which suggest leakages of radon are shown in Fig. 8. The right-hand autoradiogram was made without exposure to a light-flash to give the optical shadow of the screen, but the outline of the screen has been sketched in. Evidence of leak is shown at the soldered junction of the tube to its cap, and was great enough to put a sensitive ionometer out of action for a day after the

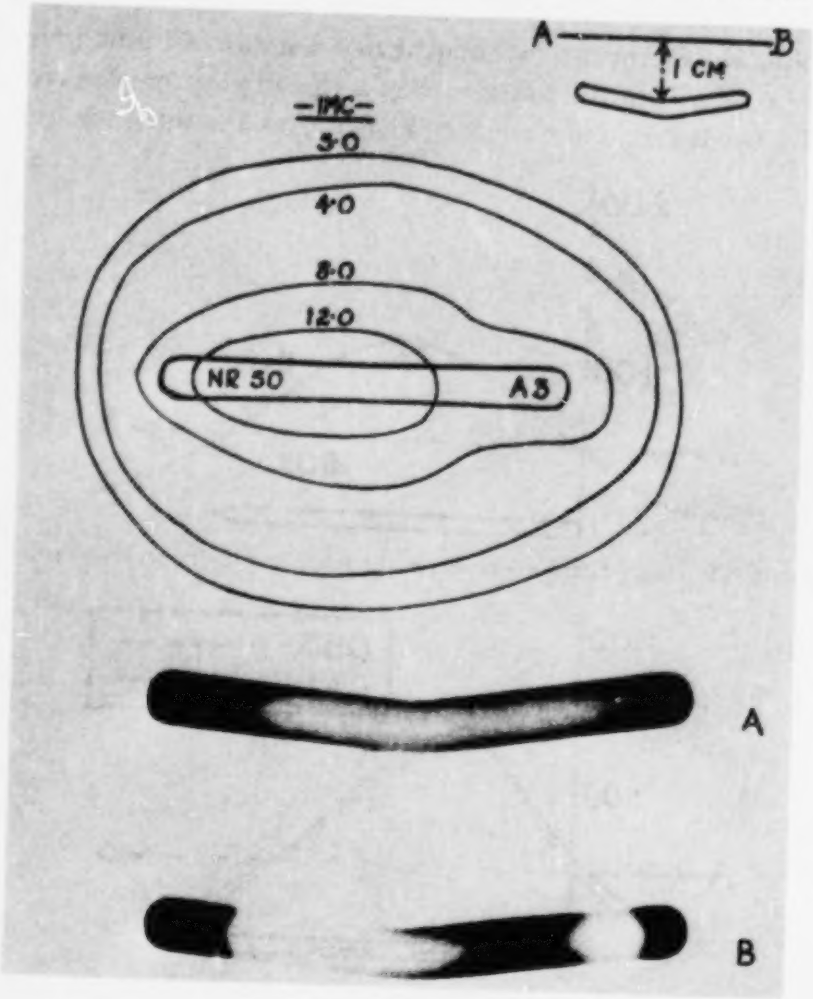


Fig. 7. Correct and badly packed intra-uterine tube, and isodose distribution near the latter.

tube in question had been left inadvertently in the laboratory for some twenty minutes. In the left-hand print, the optical shadow of the needle shaft can be seen; the shaft had been weakened where the identification numbers were too deeply engraved. This was an old, very badly filled needle with its radium content not packed in cells, the whole charge lying outside the portion shown in the figure.

Some investigations have been made of the extent to which tissue-dosage is influenced by faulty packing of needles. Trouble may be anticipated from two directions. (*a*) The dosage applied to tissue immediately in contact with the needle surface, which is usually large enough to cause necrotic change even when well packed needles are employed, will be still larger where bad distribution has resulted in abnormally great linear activity. (*b*) Discrepancies will exist between actual tissue-dose at points remote from the needle or needles, and the expected dose as calculated for an assumed standard packing.

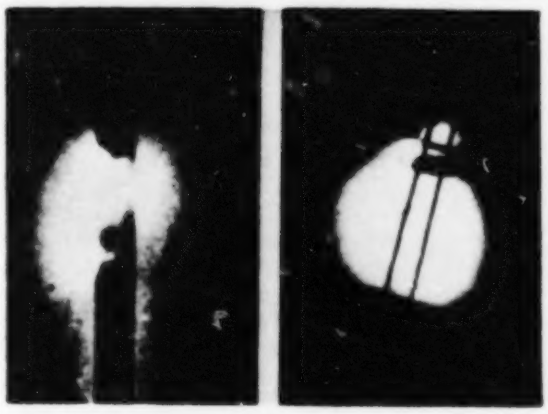


FIG. 8. Appearance suggesting leaks in needles.

(*a*) Values of the gamma-ray intensities at the surface of radium containers may be calculated from Sievert's tables:[3] as Sievert has shown, the effect of β radiation from the needle surface will be to increase the absolute effective intensity above the value obtained by calculation, but correct relative values of surface intensity when the linear activity is varied, screenage and needle dimensions remaining unchanged, may be expected from the Sievert formulæ. From the work of Benner and Snellman[4] it would seem that the effect of secondary radiation is in any case small, even at distances as small as 0·5 mm. from the needle surface. If the case of the needle of Fig. 3 is analysed along these lines, assuming the distribution found from the autoradiogram, the maximum surface intensity is found to be 25·0 Imc., whereas in the case of a correctly packed needle the value found is 13·6 Imc. At 4 mm. from the axis, the corresponding values are 5·2 and 3·05 Imc.

Table I gives data, calculated by Sievert's method, from which an estimate may be made of the volume of tissue surrounding a needle track which is subjected to excessive γ-ray intensities, and shows how this volume is increased by the abnormally high linear activity found in badly packed needles. The table gives the volume of tissue (after subtraction of the volume of the platinum needle) enclosed in the isodose surfaces for 5 Imc., 10 Imc. and 20 Imc., in the case of needles of content 2 mgm., screenage 0·5 mm. platinum, the active lengths being 2·0 cm. in case A, and 1·0 cm. in case B; the limiting case of a 2 mgm. point source, with similar screenage, is also included (case C). The diameter of the largest cross section of the isodose surfaces is also given. Cases A and B are similar to the cases of Figs. 3 and 4 in that the ratio of linear activities involved is two to one. The effect of increased linear activity on the volume of tissue which receives severe over-dosage is marked, and arises partly from the more nearly spherical shape of the isodose surfaces, and partly from the emergence into tissue of high-intensity surfaces which lie entirely inside the platinum of needles of lower linear activity.

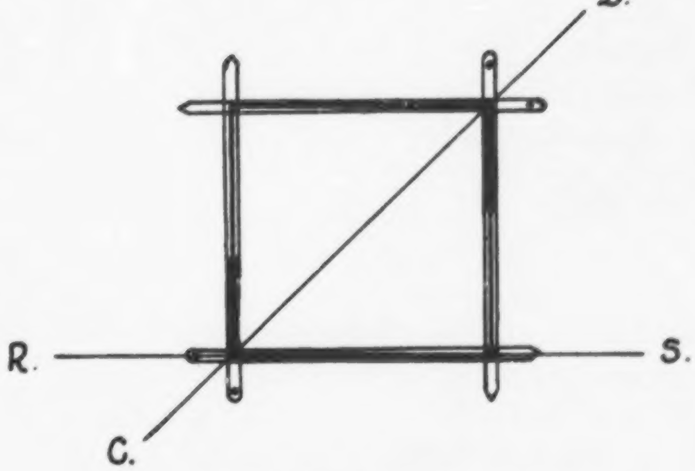
TABLE I

	Case A		Case B		Case C	
Imc.	Vol. cu. mm.	Diam. mm.	Vol. cu. mm.	Diam. mm.	Vol. cu. mm.	Diam. mm.
20	25	2·4	75	4·2	131	6·3
10	201	4·6	267	7·0	373	9·0
5	780	8·4	822	10·6	1040	12·6

Influence of linear activity on maximum diameter of isodose surfaces and on volume of tissue enclosed by them.

If it be assumed that a limit is set to the allowable time of irradiation by the necessity to avoid excessive necrosis of areas of tumour bed in the vicinity of the needle track, it is clear that a defectively packed needle will demand withdrawal substantially earlier than a correctly packed one, and that unnecessary difficulty will arise in securing adequate dosage at points more remote from the needle.

(*b*) When considerable numbers of needles are used interstitially, it is clear that the effects of non-uniformity of packing will tend to average out, and, as Quimby has shown, if the sources can be implanted in the tumour tissue, it is justifiable to base a dosage system simply on the volume to be irradiated and the total mass of radium used, distribution being variable within wide limits: but it frequently happens that groups of three or four needles only are inserted, and therefore an investigation was made as to how far bad packing might falsify the anticipated tissue dosage in such a case. Two similar plaques were made,



Plaque of four needles.

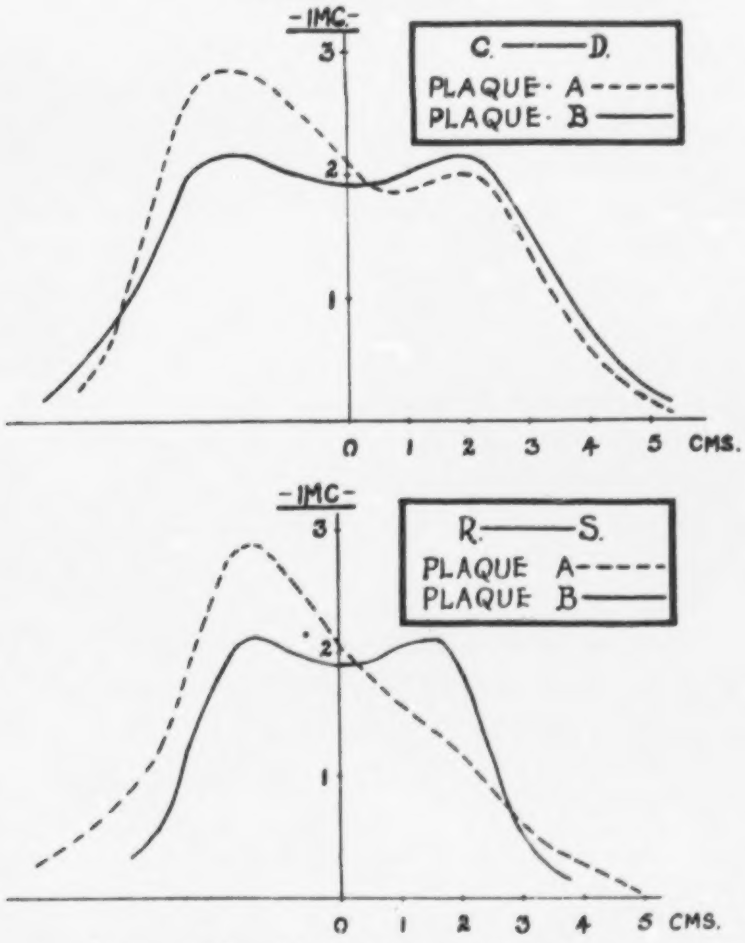


FIG. 9. Effect of bad packing on simple plaque.

using four needles in a square pattern as in Fig. 9. In plaque A, badly packed needles were employed, the radium charge being located mainly in the portions shown blackened in the figure: plaque B was made from good needles: all eight needles were specified as 2 mgm. Ra, 44 mm. total length, 0·8 mm. Pt screenage, and the total charge was correct in each needle. The γ-ray intensities were measured with the ionisation chamber of volume 40 cu. mm. at points 1 cm. above the plane of the points of contact of the needles, along the lines CD and RS of the diagram. The needle pattern, satisfactory when constructed from good needles, is seen to give dangerously non-uniform irradiation in case A. Measurements at 2 cm. from the needles showed that at this distance the effect of bad packing was present, but insignificant.

It should be emphasised that all the needles described, except those illustrated in Fig. 8, are recently-purchased needles of modern construction, in which the uniformity of distribution is supposed to be guaranteed by packing the radium salt in cells. We have observed many far worse cases among older preparations in which the radium salt is packed directly into the cavity of the shaft; in some of these the radium lay loose, and could be shaken about from end to end of the container. The discussion of intensity distributions given above has been limited to sources of modern specification, and shows that many needles are in common use in which the distribution of radium salt is so far from uniform as to give dosage distribution substantially different from that anticipated on the assumption of uniform packing; in one case demonstrated, the intensity at the needle surface was of the order of twice that to be expected, and was therefore liable to increase difficulties arising from local over-dosage; in a second case, serious regions of under-dosage were found, extending to beyond 1 cm. from a simple pattern of four defective needles. The question of distribution of radium salt within needles and applicators should, therefore, receive the close attention of all workers who wish to make a study of radium dosage along quantitative lines.

SUMMARY

Some examples of badly packed radium preparations are discussed. The magnitude of the resulting irregularities of tissue dose has been investigated, and compared with results computed from quantitative study of autoradiograms.

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A Method for Testing Radium Containers for Leakage Using the Well-Type Scintillation Counter[1]

JOHN HALE, Ph.D.

A new method for testing for leakage of radon from clinical radium containers has been developed. Preliminary tests show the method to be reproducible and highly sensitive. It is based on trapping the radon which escapes from faulty radium containers in a mixture of water and activated charcoal and subsequently measuring the radon in a well-type scintillation counter.

Method: Mix 5 ml. of tap water with 0.37 gm. (1 level 1/4 teaspoonful) of activated charcoal in a test tube suitable for well-type scintillation counter use. Place the radium container to be tested in the test tube. Stopper test tube. Leave radium in place for twenty-four hours. Remove radium. Replace stopper. Wait twenty-four hours and count in a well-type scintillation counter.

Theory: If it is assumed that radon leaks from a faulty radium container at a constant rate, the rate of leakage, *Rn*, is related to the count rate, C, by:

$$Rn = \frac{\lambda C K e^{\lambda T}}{(1-f)(1-e^{-\lambda t})} \mu c/\text{min},$$

where K (μc/count per min.) is the counter calibration factor; t is the time the radium container was left in the test tube; T is the time interval between removal of radium and counting; λ is the radon decay constant; and f is the fraction of radon lost when the radium is removed from the test tube.

Experimental: To establish the values of the constants involved in the equation, preliminary tests were made on 5–0.5-mg. radium cells previously known to be leaking. The half-life of the radioactivity within the test tube was observed to be 3.9 ± 0.1 days, confirming that the radioisotope was radon in equilibrium with its decay products. Leakage through the stopper was less than 1 per cent per day. Additional tests showed that f was less than 1/50. Repetitive tests on the same radium containers showed that the method was reproducible to about ±7 per cent. The counting sensitivity for a conventional well-type scintillation counter was found to be 1.2×10^6 counts per minute per μg. by comparison with radium standards. Under the conditions given above, and for a standard error of 50 per cent of net count rate with a one-minute sample count and a ten-minute background count, the minimum detectable leak was 2.5×10^{-8} mc for the twenty-four hours of the test. This corresponds to 1.4×10^{-7} mg. of completely de-emanated radium.

Conclusions: The method appears to have the following advantages:

1. With proper planning of operations, a large number of radium containers can be tested easily.
2. Handling procedures are simple, so that exposure to the operator is minimal.
3. The method is quantitative.
4. The method is quite sensitive, ample to detect significant radon leakage from clinical sources.
5. Previous methods used for testing clinical radium containers have been based on the measurement of alpha particles from Ra A-B-C (effective half-life about twenty minutes), so that counting had to be done immediately. The measurement of radon, to the contrary, allows counting to be done several days after testing. This makes it possible to use a central counting facility for radiation safety survey work, since the radium can be tested in its normal storage place and only the test tubes transported to the counting facility.

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[1] From the Department of Radiology, Hospital of the University of Pennsylvania and the Wm. H. Donner Center for Radiology, University of Pennsylvania, Philadelphia 4, Penna. This work was supported in part by a grant (C-1984) from the National Cancer Institute, U. S. Public Health Service.

INSTRUMENTAL AND TECHNICAL NOTE

THE EXAMINATION OF RADIUM CONTAINERS FOR RADON LEAKS

By A. F. Holloway, Ph.D., and C. J. Mandy

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If radon is detectable in the air surrounding a radium source, it must have escaped from the container through some imperfection, or be due to contamination of the outside of the sealed source. If the leak is very large, a substantial proportion of the RaC and other daughter isotopes will be deposited at some place other than the supposed source. Thus the leaking tube will not be giving off the expected amount of γ rays and dosage to patients will be lower than planned. A leak of this size is very rare, but easily detected when it does occur by the simple cotton wool and geiger counter test.

A more common variety of leak is one which produces only a low concentration of radon in the air surrounding the source. While such a leak does not affect the exposure dose delivered by the source at a given distance to any measurable extent, it nevertheless indicates an imperfection in the container. This imperfection suggests a weakness in the sheath which should be repaired before additional damage can cause a radium spill. These small leaks are not readily detectable by conventional tests but are easily found with an ionization chamber filled with air from a jar which has been sealed overnight with the source inside (Turner, private communication).

The equipment used in the present investigation consists of a brass ionization chamber of 1·22 l. internal volume. The ionization current is measured with a Townsend balance circuit, using a Lindemann electrometer. The background current with this arrangement is from 1 to 2×10^{-15} amp and the current with the maximum permissible concentration of 10^{-10} c of radon per litre of air is 2×10^{-13} amp. In use the ionization chamber is evacuated and sealed with a stop-cock. It is then moved to a region of interest, the stop-cock is opened while a sample of air is sucked into the chamber and then closed again. The ionization current in the chamber may now be measured. If any activity is present, accurate measurements must be deferred for three hours to allow the decay products of radon to come to equilibrium with the radon present in the chamber. The identity of the radioactive contaminant may be checked by half-life measurements. Several samples of air were taken from the neighbourhood of the radium safe. Later the radium containers were checked by sealing overnight in a jar and then sucking the air from the jar into the ionization chamber. Any leaks were traced to individual tubes or needles and these were isolated None of the leaks found was detectable by the cotton wool and geiger counter technique.

The radon concentration from the vicinity of the safe was found to be less than 10^{-12} c per litre of air, which is less than 1 per cent of the maximum permissible level for occupational exposure.

Of the 97 radium containers investigated only four were found to have leaks and these were all small. The largest leak, from a 10 mg needle with 0·6 mm platinum walls, produced 65×10^{-12} c of radon when sealed in a jar for 16 hours. Because the radium was only sealed for this short time this represents only 11·4 per cent of the equilibrium amount of radon that would have been present after an "indefinitely long" (more than 30 days) sealing period. The others, after comparable times, produced 13×10^{-12} c, 6×10^{-12} c and 32×10^{-12} c of radon respectively.

The first three leaks all occurred in needles that were sealed in 1934 and the leak, considered in conjunction with the age of the needles gives strong evidence that they should be resheathed. The last leak mentioned was found in a needle only one and a half years old, but it is such a definite leak that it indicates some defect in the seal or possibly is evidence of damage, although superficial inspection does not indicate that this is likely.

The question of a useful life for a container which has not been damaged exteriorly is complicated by the possibility that large gas pressures due to helium produced in the decay may develop. Atomic Energy Co. of Canada, who have opened many old radium tubes, were asked whether they had ever observed any effects attributable to such pressures and also what they would recommend as a useful life for the containers. Their answer (Beach, private communication) was that their "laboratory has checked needles and tubes that have been in use for 15 and 20 years and in every case the gas pressure was negligible". They had found, however, that the solder gets weak with age and that the platinum walls appear to become porous to radon gas.

Acknowledgment

The authors wish to express their gratitude to Mr. J. G. Beach of Atomic Energy of Canada for his advice and permission to publish excerpts from his letter.

(A.F.H. is now at The Manitoba Cancer Treatment and Research Foundation, Winnipeg, Manitoba.)

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METHODS USED FOR TESTING RADIATION SOURCES FOR LEAKAGE AND CONTAMINATION

A. Wipe test for alpha sources.

All surfaces of the source (excepting the cut ends of foils) are wiped gently with a tissue moistened with a volatile solvent. The activity of the swab is counted with a scintillation counter.
Limit: 3 c.p.s. above background, corresponding to < 0·01 μc.

B. Wipe test for beta sources.

All surfaces of the source are wiped with a tissue moistened with a volatile solvent. The activity of the swab is measured with an end window counter shielded if necessary by a lead castle. This is similar to the wipe test in Appendix A of B.S. 3513: 1962.
Limit: 0·05 μc; normal level < 0·001 μc.

C. Wipe test for gamma sources.

The source is rubbed into a moistened swab or tissue, and the activity measured with an end window counter. This test is similar to the wipe test in Appendix A of B.S.3513:1962.
Limit: 0·05 μc; normal level < 0·001 μc.

D. Bubble test.

The source is immersed in a suitable liquid (diethanediol) and the pressure in the vessel reduced to 100 mm of mercury. No bubbles should be observed. This test conforms to the requirements of B.S. 3513: 1962.

E. Emanation test.

The source is placed in a glass specimen tube with a length of polythene tubing (40 × 3mm); the tube is closed by a rubber bung and left for 48 hours. The beta activity of the polythene is measured with an end window counter.
Limit: 5 c.p.m. above background.
(It is difficult to assess to what this rate corresponds, but an estimate is 7×10^{-9} c radon in 12 hours.)

F. Immersion test for beta and gamma sources.

The source is immersed in water at 50°C for 8 hours and the activity in the water measured. This is the immersion test stated in Appendix B of B.S. 3513: 1962.
Limit: 0·05 μc.
For sources intended for use in the U.S.A. the test is repeated after 30 days; limit 0·005 μc.

G. Immersion test for caesium-137 sources.

The source is immersed in water which is raised to boiling point and held for 5 min. The water is then removed and tested for activity. The source is cooled and the test repeated twice. Sources are passed if the total extracted in two consecutive tests does not exceed 0·05μc, and if successive tests do not show a rise in the activity detected.

H. Helium mass spectrograph test.

This test is used for welded sources.
Limit: leak rate of 10^{-6} standard cc/sec.

K. Emanation test (Scintillation counting test for radon).

The appliance is immersed in a solution of a phosphor in an organic liquid under vacuum; the leakage of radon is measured by liquid scintillation counting.
Limit: about 5×10^{-11}c per 24 hours.

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SIMPLE TEST FOR LEAKING RADIUM SOURCES*

By JOHN E. ROSE, D.Sc.
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AN EXTREMELY sensitive, reliable and inexpensive test that even the inexperienced may use with confidence has been available for over 30 years which may be of help to institutions and radiologists faced with the task of routinely testing their sealed radium sources. The procedure requires little manipulation, can be done quickly with simple forceps and with negligible exposure to the operator.

The tube, plaque, needle, etc., is placed in the bottom of a clean test tube or screw-top glass bottle, being careful not to contaminate the sides or top in case the preparation is broken and leaking salt. A container which will comfortably accept the preparation and at the same time accommodate a loosely packed ball or wad of ordinary surgical cotton with a volume of 2 or 3 cubic inches is chosen. The cotton is suspended or packed in such a way as to ensure that it does not actually come in contact with the source. The container is closed overnight with a stopper or screw-top lid; no special seal or gasket is required.

The next morning, or at some convenient time about 16 or more hours later (little is to be gained beyond 2 days), the cotton is removed and held next to the 30 mg./cm.^{2} window of a portable Geiger-Müller survey meter. With earphones, any increase over background indicates a leak of the order of 10^{-9} curies. About 10^{-8} curies will kick the meter off scale yielding thousands of counts per minute, more or less. If the radiologist or the institution does not own a portable Geiger-Müller counter, one might be borrowed from the local Civil Defense Office or a consulting physicist.

In explanation, the object of the cotton is to introduce into the test container the largest possible surface without contamination upon which most of the active deposit from the decaying radon collects. In removing the cotton one transports the active deposit (Ra B and C with a half-life of about 27 minutes) and its energetic betas to the Geiger-Müller tube. If the cotton occupies most of the volume of the container, the sensitivity is at its highest.

For grossly leaking preparations one needs only to place cotton in the same drawer with the suspect preparation in the radium safe overnight or in some cases for a few minutes.

The inexperienced may practice on his own radium dial wrist watch (after loosening the face) since most dials yield 0.1 μc or more radioactivity.

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DAILY LEAK CHECKING OF THE ENTIRE STOCK IN A RADIUM SAFE*

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*Presented at the Tenth Annual Meeting of the Health Physics Society, Los Angeles, California, 17 June 1965.

†Operated by the University of Chicago for the United States Atomic Energy Commission.

DAILY LEAK CHECKING OF THE ENTIRE STOCK IN A RADIUM SAFE

As a precaution against the hazards associated with the dispersal of radium salts, including personnel exposures and decontamination costs, it is common practice to test periodically for the presence of holes or fractures in radium needles and radium tubes used for radiation therapy. Such testing is mandatory in some states.

Radon, present as the gaseous daughter product of radium, tends to escape from needles through holes too small for the passage of radium salts. The detection of the escaping radon forms the basis of commonly accepted leak testing procedures.

We have found, however, that holes in radium needles and tubes can be temporarily plugged with water thus preventing the escape of radon and thereby rendering the holes undetectable. Since it is possible that some needles might be plugged with water during a periodic leak test and later dry out and begin leaking again, it is obvious that more frequent leak testing is required. Heretofore leak tests were performed at six-month intervals; we have reduced the interval to one day. Daily testing of the entire radium stock is performed automatically by the monitoring system to be described below.

First, we will present our observations of two commonly used leak testing procedures and compare them with a procedure we found desirable. Second, we will describe our experience in plugging and unplugging leaky radium needles, and finally we will present a description and performance data of our automatic monitoring system.

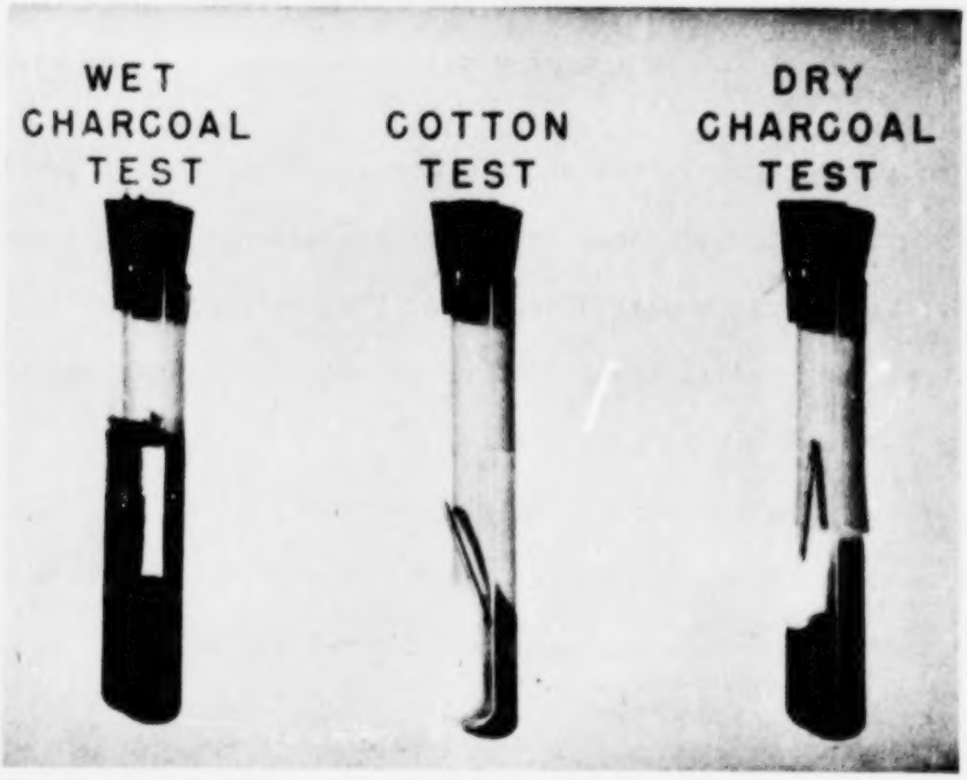

Figure 1. Three methods used to test for radon leaks showing needles in place: wet charcoal, cotton, and dry charcoal.

Figure 1 shows the wet charcoal test,[1] the widely used cotton test, and a dry charcoal test which we devised in the course of our work. In all three tests the radium needle is left in the stoppered test tube for 24 hours. The needle is then removed, the test tube stoppered again, and counted in a well type scintillation counter.

The wet charcoal test was found to plug up needles with large radon leaks. Since it could not be recommended for leak testing, no further investigation was made of this procedure. Figure 2 shows the distribution of activity in the other two tests, cotton and dry charcoal, as measured with a collimated detector. More activity is in the stopper end than in the cotton end of the cotton test while most of the activity in the dry charcoal test was retained on the charcoal. Thus a substantial

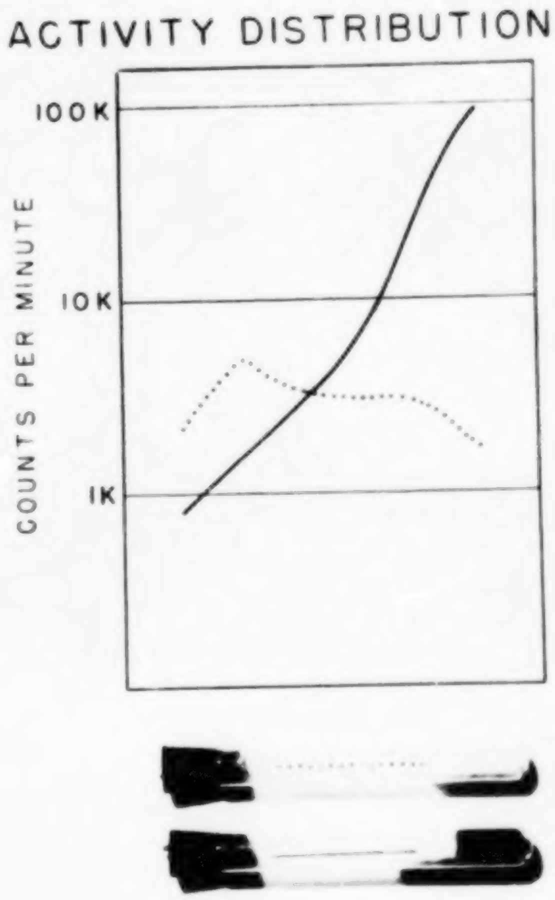

Figure 2. Distribution of radon activity in the two dry leak tests. The charcoal concentrates the radon while the cotton does not.

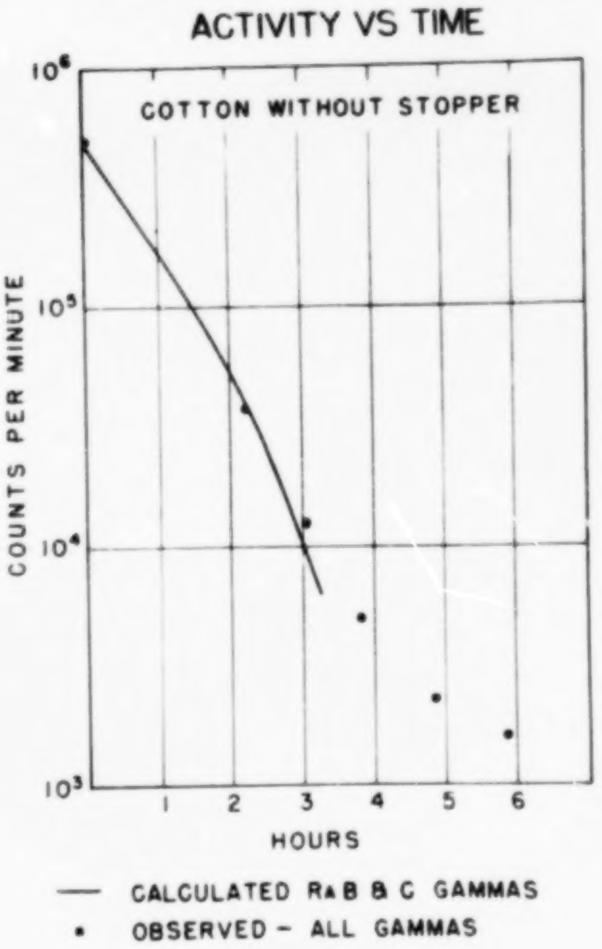

Figure 3. Comparison of theoretical and observed decay of radon daughters showing the rapid diffusion of radon out of the cotton after removal of the stopper.

geometrical correction factor is required for the cotton test if quantitative counting results are to be obtained, while no such factor is required for the dry charcoal test.

The activity in the bottom end of the cotton test fell off rapidly after the stopper was removed, Figure 3 shows a comparison of the data points with the Bateman equations assuming that all the radon instantly diffused out of the test tube and all the daughters were trapped in the

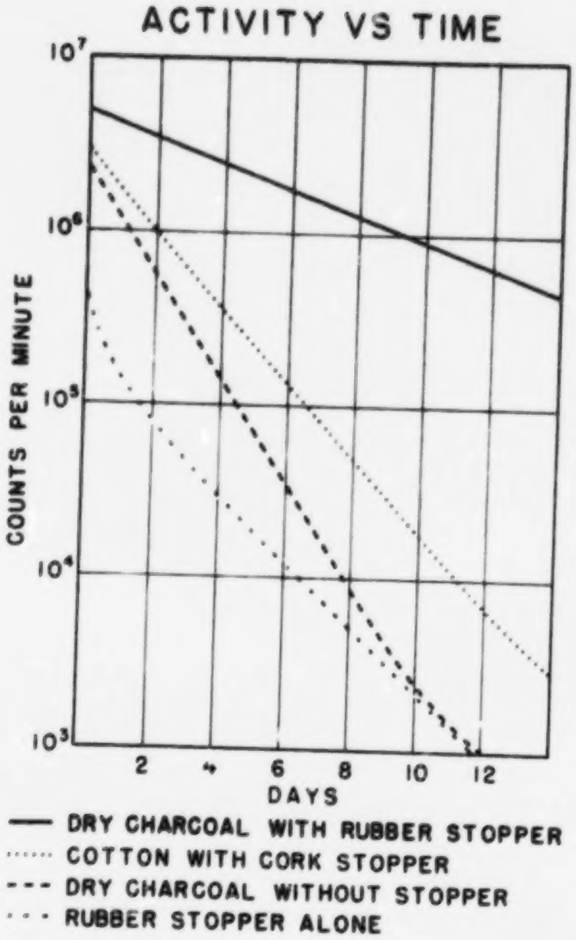

Figure 4. Decay of activity measured over a period of several days, showing the radioactive decay in the dry charcoal test, combined radioactive decay and diffusion from the charcoal, combined decay and diffusion through a cork stopper, and out of a rubber stopper.

cotton. We were careful to keep the test tube vertical during this six-hour period so that we observed true diffusion of the radon and not simply a "pouring" out of the heavy gas. The activity in the bottom of the dry charcoal test tube declined at a much slower rate after removal of the stopper as shown in Figure 4. Also plotted, for comparison, are the decline in counts from a dry charcoal test with rubber stopper in place, from a cotton test with a cork stopper and from a rubber stopper alone.

We thus conclude that the dry charcoal test with rubber stopper is the best of the three studied. There is no water to plug holes, it retains the activity at the bottom of the test tube thereby requiring no geometrical correction, and only a negligible amount of radon is lost during the few seconds required to remove the radium needles. Cork stoppers should be avoided since they are porous to radon.

The tests just described were used, with normalizing corrections, to investigate the variation of radon leak rates from five doubly encapsulated radium needles and tubes known to have holes. Figure 5 shows that, during the last week in January, needle No. 6 stopped leaking each time it was

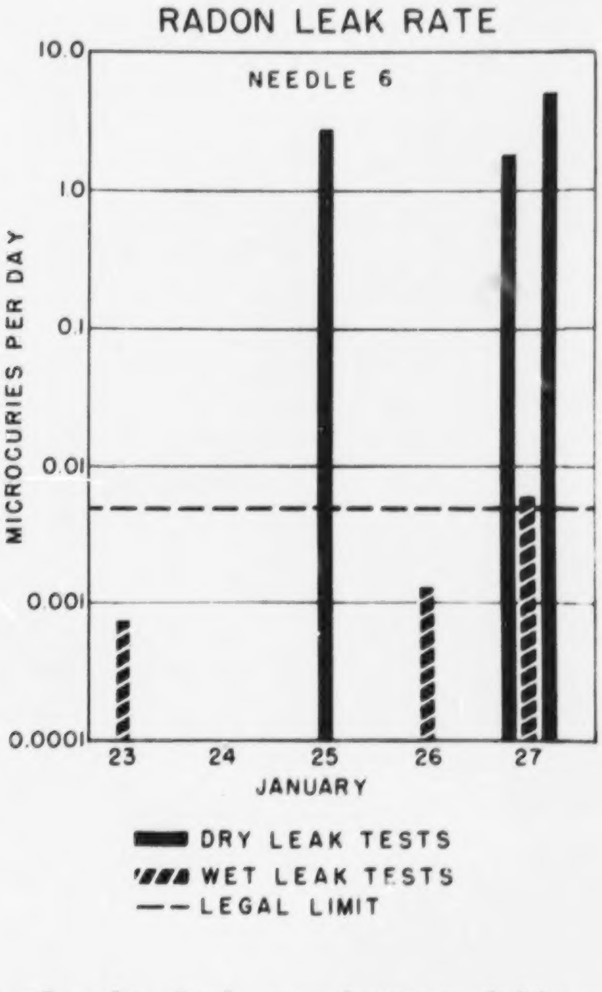

Figure 5. Variation of radon leak rate between daily wet and dry leak tests, showing that water consistently plugged the leak in needle No. 6.

subjected to the wet charcoal test, i.e., when submerged in water, and began leaking again later when dry. It should be noted that Figure 5 is a semi-log plot and the ratio of dry to wet leak rates is about one thousand. The dashed line at 0.005 microcuries represents our interpretation of the legal limit for removable contamination when applied to radon leaks. The last three bars crowded together on January 27, represent averages of several one-half hour tests made that day. The results of these individual one-half hour tests are shown in Figure 6. It is apparent from Figure 6 that this needle stopped leaking soon after

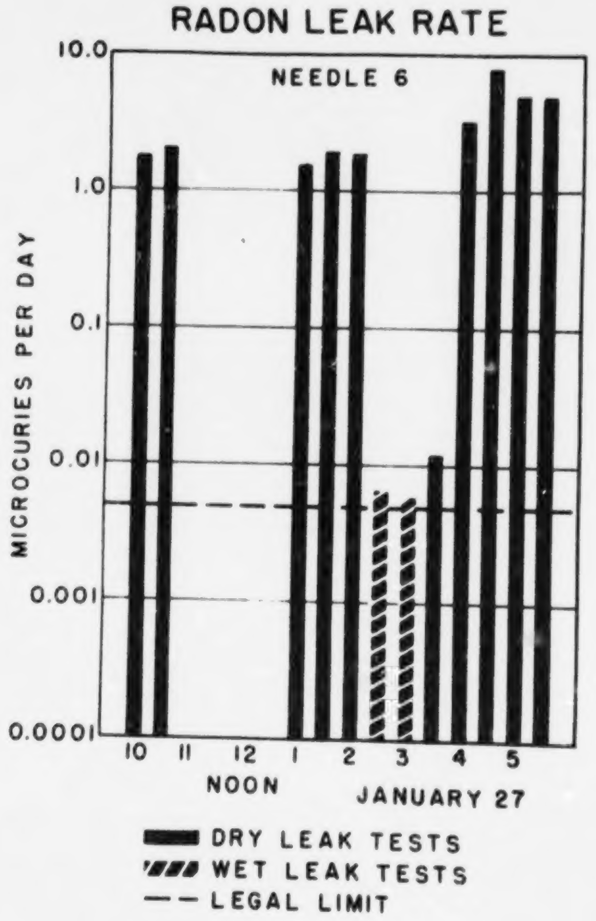

Figure 6. Variation of radon leak rate between half-hour wet and dry leak tests, showing that the leak remained plugged for some time after removal from water.

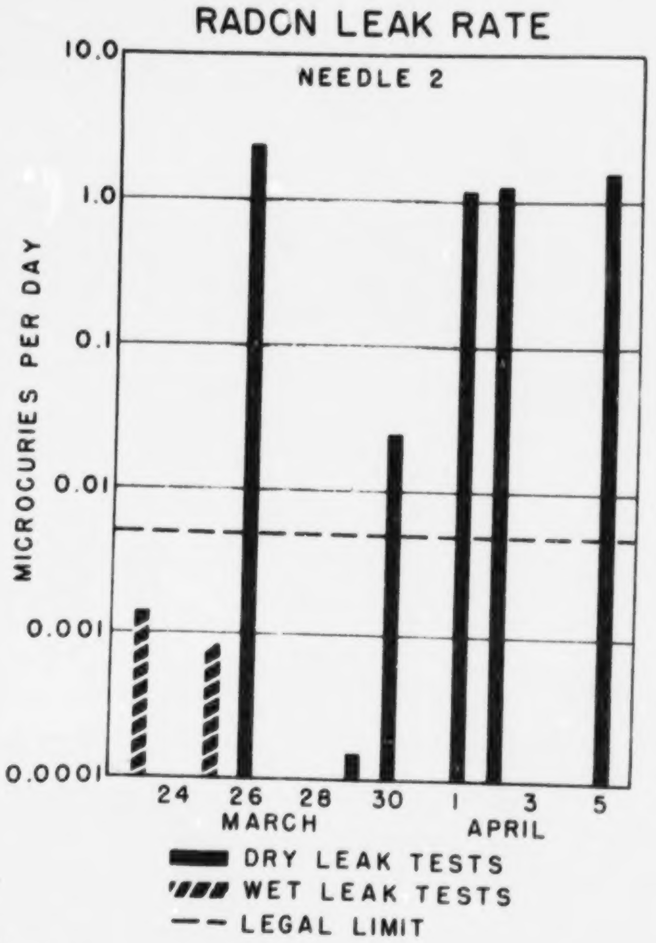

Figure 7. Daily tests on needle No. 2 show a temporary plugging for several days after removal from water.

immersion in water and, in this instance, took one-half hour to dry out and begin leaking again. The actual activity accumulated at the end of these one-half hour tests was multiplied by the appropriate factor to give the activity that would have accumulated at the end of one day tests.

Figure 7 shows that needle No. 2 also became plugged when wet and unplugged when dry. This time, however, the needle became temporarily plugged again on March 29, even though it was kept dry.

We see in Figure 8 that needle No. 6 also became plugged again on March 30 even though it was kept dry. This time, however, it remained plugged for several weeks. We then heated the needle to 100°C in charcoal in an effort to evaporate the water that had plugged the leak. Due to insufficient dehydration of the charcoal, the needle received a steam bath and either the heat, or steam, or both sealed the needle so that the radon leak rate fell below the minimum detectable activity of our equipment. Several additional 100°C heatings with adequately dehydrated charcoal failed to unplug the hole. Finally, after heating in dry charcoal for several hours at 150°C the hole was unplugged, and after an initial burst of radon probably due to built up pressure, the leak rate returned to approximately the level of two months earlier (see Figure 5).

Three additional needles and tubes which had been leaking at consistent measurable rates were individually subjected to 150°C heat in dehydrated charcoal. Two needles did not respond to treatment but radium tube No. 5, after an initial surge of radon and a visible amount of water, leaked consistently above the legal limit as shown in Figure 9.

The conditions that these needles were subjected to are not uncommon. Needles are placed in a liquid for cold sterilization, or washing. Body

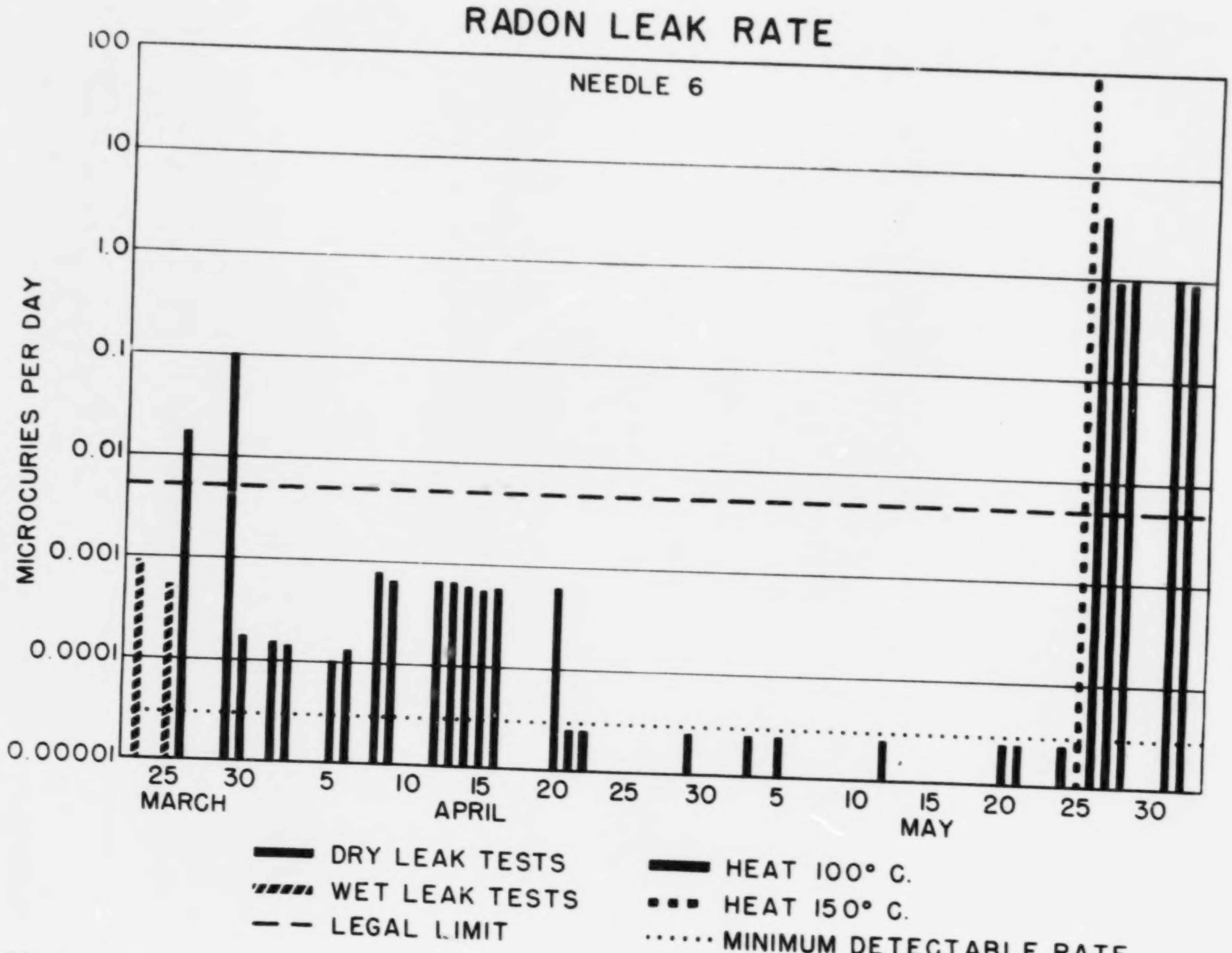

Figure 8. Two-month history of leak rate of needle No. 6. Water plugged this needle for an extended period of time and heating was required to unplug the leak. Leak then rose to its previous level (see Figure 5)

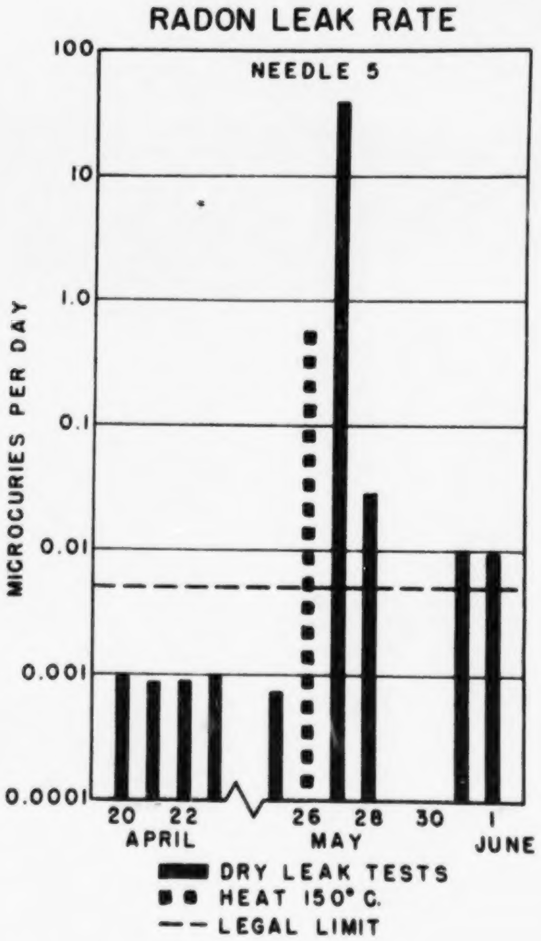

Figure 9. Daily tests on needle No. 5 showing effect of heating on needle that had leaked at a low rate for some time. Water was ejected from this needle by the heating process.

fluids from the patient could also plug the leaks. Some installations boil their needles, others autoclave them, and they are sometimes heated in the preparation of molds. Thus it is quite likely that there are a number of such faulty, but plugged, needles lying undetected in many radium safes.

We do not recommend the potentially dangerous procedure of heating entire stocks of needles and tubes to 150°C, because the internal pressure is increased by 43% over the pressure at room temperature and this might cause some needles to explode.[2] At the present time we prefer to wait until the needles dry out, and detect the leak whenever it occurs by monitoring the entire stock every day. This is accomplished by

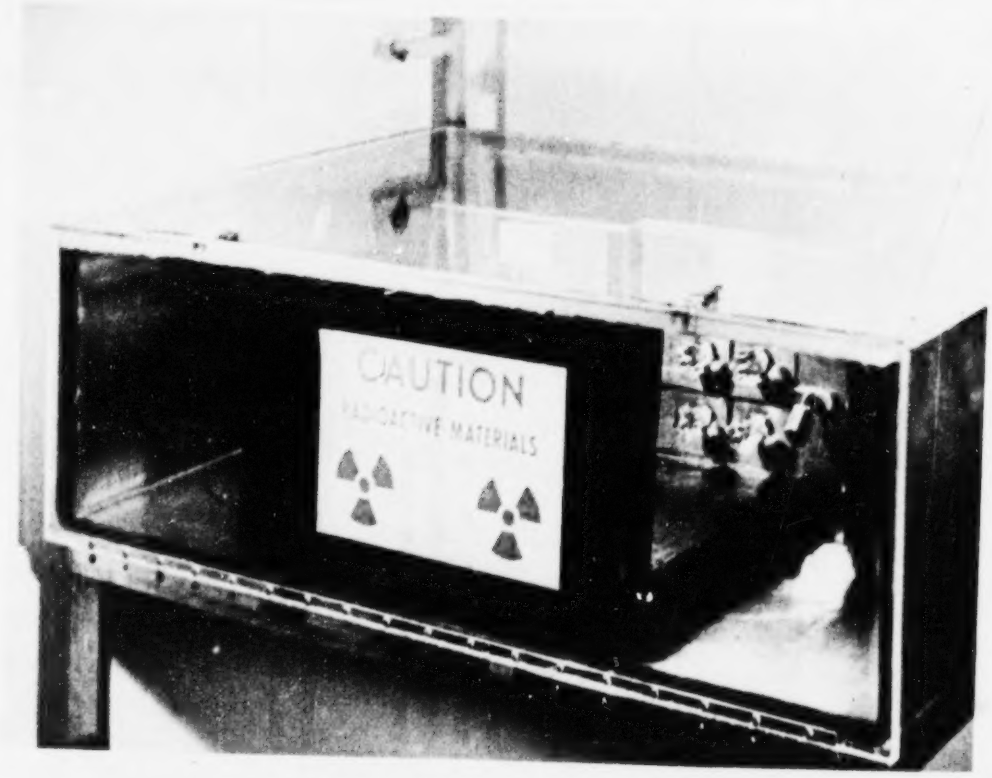
CAUTION
RADIOACTIVE MATERIALS

Figure 10. The air sampling enclosure containing the radium safe.

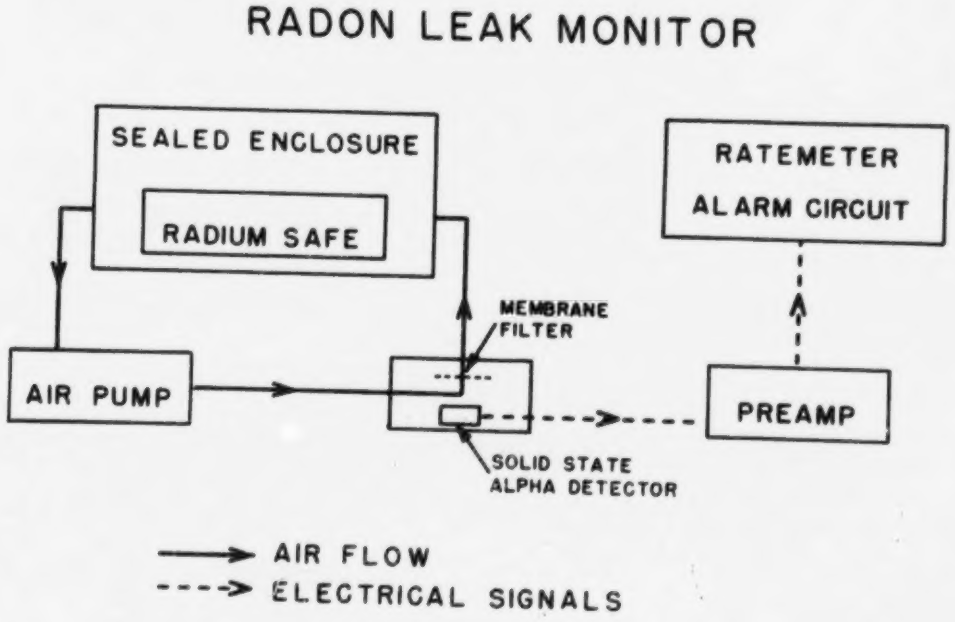
RADON LEAK MONITOR
SEALED ENCLOSURE
RADIUM SAFE
RATEMETER
ALARM CIRCUIT
MEMBRANE FILTER
AIR PUMP
PREAMP
SOLID STATE ALPHA DETECTOR
AIR FLOW
ELECTRICAL SIGNALS

Figure 11. Block diagram of radon leak monitor, based on automatic air sampling technique.

mounting the radium safe in an air tight enclosure as shown in Figure 10. The top and front of the enclosure can be opened wide to allow free access to the safe, but they are kept closed at all other times.

The air in the enclosure is pumped daily in a closed circuit through a membrane filter as shown in Figure 11. Radon daughters, if any are present, are deposited on the filter and counted with a silicon surface barrier detector. This type of detector was chosen because its thin depletion region gives a low gackground count rate in the presence of gamma radiation near the safe. Sequencing is accomplished automatically by a timer and visual alarms are designed to be fail safe.

Several factors had to be considered in order to achieve adequate sensitivity: filter pore size, air flow-rate, sampling time, and interval between sampling. Pore size must be small enough to collect a substantial fraction of the dust to which the radon daughters attach themselves; too small pores, however, will restrict the air flow which must be great enough to collect a substantial number of radium A atoms in a time that is short compared to its half life. Sampling time greater than a few minutes does not increase collection of radon daughters because the dust concentration is lowered. The enclosure should remain closed for the longest possible time before sampling in order to increase the radon concentration and thereby the sensitivity of the test.

The system described, using 0.45 micron pore size and 60 lpm air flow-rate, does not as yet operate at its optimum efficiency. Sensitivity of this system is 200 cpm per 0.005 microcuries of radon in the enclosure, as determined by calibrating the leak rate of the leaking needles with a 0.1 microgram NBS radium standard in a NaI(Tl) well counter.

Finally, small installations with a few needles can test more economically by storing them in sealed test tubes with dry charcoal. When the needles are removed for use, but prior to use, the test tube may be counted with a GM or equivalent instrument. The instrument must be selected with care; one 40 mg/cm^2 aluminum walled Geiger tube was observed to have a minimum detectable activity of 0.0005 microcuries of radon daughters, while a 3.5 mg/cm^2 end window Geiger tube was much less sensitive and had a minimum detectable activity of 0.014 microcuries.

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1. Hale, John: A Method for Testing Radium Containers for Leakage Using the Well-Type Scintillation Counter, Radiology. 72: 105. January, 1959.
2. Morgan, J. E.: Gas Pressure in Sealed Radium Containers. The American Journal of Roentgenology, Radium Therapy and Nuclear Medicine. 85: 949-954. May, 1961.

AN APPROACH TO LEAK TESTING OF SEALED RADIUM SOURCES*

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AN APPROACH TO LEAK TESTING OF SEALED RADIUM SOURCES

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Introduction

There are several different methods for leak testing sealed radium sources. These methods can be classified into two groups:

1) Those which collect radon plus the radon daughters

2) Those which collect only the daughters of radon

There are three factors to be accounted for in all of these methods; a) the length of collection time, b) the elapsed time between end of collection and counting, and c) the type of radiation (alpha, beta, or gamma) to be counted.

In this paper two approaches are presented to relate data from various leak test methods with allowances for the above mentioned factors. Therefore, with knowledge of the general leak test method (1 or 2) and the three variable factors (a,b, and c), instantaneous alpha or beta counts can be extrapolated back to total counts associated with the original radium which created the "leak test" counts.

Instantaneous Count Extrapolation Factors

Two extrapolation factors are calculated for each of the two leakage determination methods. The first extrapolates an instantaneous alpha count back to the total counts associated with the original radium which created this activity, and the second extrapolates an instantaneous beta count in the same manner.

Generalized Bateman Equation

Since calculations are somewhat lengthy, it is helpful to introduce a compact and generalized form of the Bateman equations.

$$f_{\frac{n}{m}}(t) = (-1)^{n-m} \left[\prod_{i=m+1}^{n} \lambda_i \right] \sum_{j=m}^{n} \left[e^{-\lambda_j t} \prod_{k=m}^{n} \frac{1}{(\lambda_j - \lambda_k)} \right]$$

with $n > m$, and $k \neq j$.

Where t represents time and the subscript of each λ indicates the position of the associated nuclide in the decay chain. (For instance, λ_1 is the decay constant of radium, λ_2 the decay constant of radon, etc.) m and n are indices related to the position of the nuclide in the decay chain. The use of m and n can readily be understood by observing that the physical interpretation of $f_{\frac{2}{1}}(t)$ is the ratio of the activity of radon relative to the activity of the pure radium when the radium was considered pure t hours ago.

Tables I-A and I-B

In the radon plus daughters system, the radon gas emanated from a leaking needle is collected. A substance such as activated charcoal is used to trap the radon. The charcoal is left in contact with the needle for a variable length of time, say x hours. The charcoal is then removed, and a period of y hours elapses before an instantaneous alpha or beta count is taken.

Evaluation of the following formula yields an extrapolation factor E_{IA} which is a function of the variables x and y. This factor, when divided into the observed alpha count, produces the counts associated with the total activity of the original radium.

$$E_{IA} = f_{\frac{2}{1}}(x)\, e^{-\lambda_2 y} + f_{\frac{2}{1}}(x)\, f_{\frac{3}{2}}(y) + f_{\frac{2}{1}}(x)\, f_{\frac{5}{2}}(y)$$
$$+ f_{\frac{3}{1}}(x)\, e^{-\lambda_3 y} + f_{\frac{3}{1}}(x)\, f_{\frac{5}{3}}(y)$$
$$+ f_{\frac{4}{1}}(x)\, f_{\frac{5}{4}}(y) + f_{\frac{5}{1}}(x)\, e^{-\lambda_5 y}$$

The corresponding extrapolation factor for the beta count is

$$E_{IB} = f_{\frac{2}{1}}(x)\, f_{\frac{4}{2}}(y) + f_{\frac{3}{1}}(x)\, f_{\frac{4}{3}}(y) + f_{\frac{4}{1}}(x)\, e^{-\lambda_4 y}$$
$$+ f_{\frac{2}{1}}(x)\, f_{\frac{5}{2}}(y) + f_{\frac{3}{1}}(x)\, f_{\frac{5}{3}}(y)$$
$$+ f_{\frac{4}{1}}(x)\, f_{\frac{5}{4}}(y) + f_{\frac{5}{1}}(x)\, e^{-\lambda_5 y}$$

E_{IA} and E_{IB} are evaluated for variable values of x and y in Tables I-A and I-B respectively.

Tables II-A and II-B

In the radon daughter system, only the radon daughters are collected. Exactly the same procedure is followed as in the previous system. The alpha count extrapolation factor is:

$$E_{IIA} = E_{IA} - f_{\frac{2}{1}}(x) \left[e^{-\lambda_2 y} + f_{\frac{3}{2}}(y) + f_{\frac{5}{2}}(y) \right]$$

The beta count extrapolation factor is:

$$E_{IIB} = E_{IB} - f_{\frac{2}{1}}(x) \left[f_{\frac{4}{2}}(y) + f_{\frac{5}{2}}(y) \right]$$

E_{IIA} and E_{IIB} are evaluated for variable values of x and y in Tables II-A and II-B respectively.

Extension of Tables

The tables which are included can be extended to any conveniently applicable x and y times by means of an available computer program for the IBM 1620 written in Fortran. The results may be plotted as shown in Figures 1 and 2.

Model Interpretation and Development

The extrapolation factor can be physically interpreted to be the ratio of either alpha or beta activity (whichever is applicable) to the total activity of the radium which produced this activity or the activity of radon at equilibrium. The alpha or beta activity is determined at time $t = x + y$, and the radium activity is determined at $t = 0$. As an example of the technique used to develop an extrapolation factor, the components of E_{IA} are listed and interpreted.

It is assumed that the original activity of the radium is unity, that the activity of the RaC equals the activity of RaC', and that the activity of the RaD is zero.

1. $f_{\frac{2}{1}}(x)\, e^{-\lambda 2^{y}}$: The radon present in the system after x hours, decayed for y hours.
2. $f_{\frac{2}{1}}(x)\, f_{\frac{3}{2}}(y)$: The RaA ingrown in y hours from the radon present in the system after x hours.
3. $f_{\frac{2}{1}}(x)\, f_{\frac{5}{2}}(y)$: The RaC' ingrown in y hours from the radon present in the system after x hours.
4. $f_{\frac{3}{1}}(x)\, e^{-\lambda 3^{y}}$: The RaA present in the system after x hours, decayed for y hours.
5. $f_{\frac{3}{1}}(x)\, f_{\frac{5}{3}}(y)$: The RaC' ingrown in y hours from the RaA present in the system after x hours.
6. $f_{\frac{4}{1}}(x)\, f_{\frac{5}{4}}(y)$: The RaC' ingrown in y hours from the RaB present in the system after x hours.
7. $f_{\frac{5}{1}}(x)\, e^{-\lambda 5^{y}}$: The RaC' present in the system after x hours, decayed for y hours.

All nuclides which are alpha emitters are systematically considered and the extrapolation factor is the sum of all the individual contributions.

EXAMPLES OF TABLE USAGE

Radon Daughter System

A radium needle is placed in an electrostatic leak testing apparatus(1) for 4 hours. The cathode is removed and counted 6 minutes later using an alpha field instrument with an overall efficiency of 50%. The count rate was 150 cpm.

What quantity of radium or radon in equilibrium can be associated with this alpha activity?

From Table II-A, $E_{IIA} = .029$ for $x = 4$ hours, $y = 0.1$ hour. Therefore:

$$\text{Radium Activity} = \frac{150 \text{ cpm}}{(.5 \text{ efficiency})(.029)\left(2.2 \frac{\text{cpm}}{\text{pc}}\right)} \approx 5{,}000 \text{ pc}$$

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(1) Gallahar, Evans, McCallister, "Testing Radium Capsules for Radon Leakage," American Journal of Roentgenology, Radium Therapy, and Nuclear Medicine: Vol. XC, no. 2, August 1963.

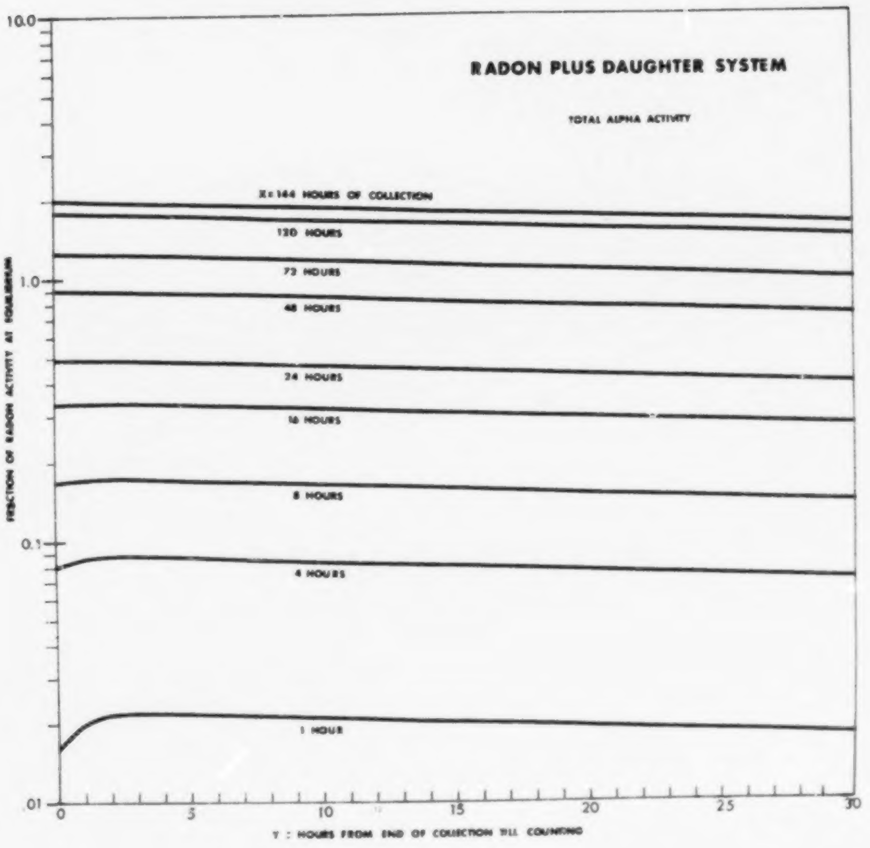

Figure 1. This is a plot of the extrapolation factor E_{IA} as a function of the time y that has elapsed from the end of leak testing till the sample was counted. The plot presents typical ingrowth or leak testing times x from 1 to 144 hours. This graph is only applicable when a radium leak test method is used that collects radon plus the radon daughter products and when the sample is counted for alpha activity. The slow rate of decay of the activity is due to the 3.83 day half life of radon in the sample. The use of this method for leak testing would require corrections for counting efficiency, absorption of the radon into the sample (e.g. charcoal), and considerations that the radon may "off-gas" from the sample after it has been collected. The plot is a graphic presentation of the data contained in Table I-A.

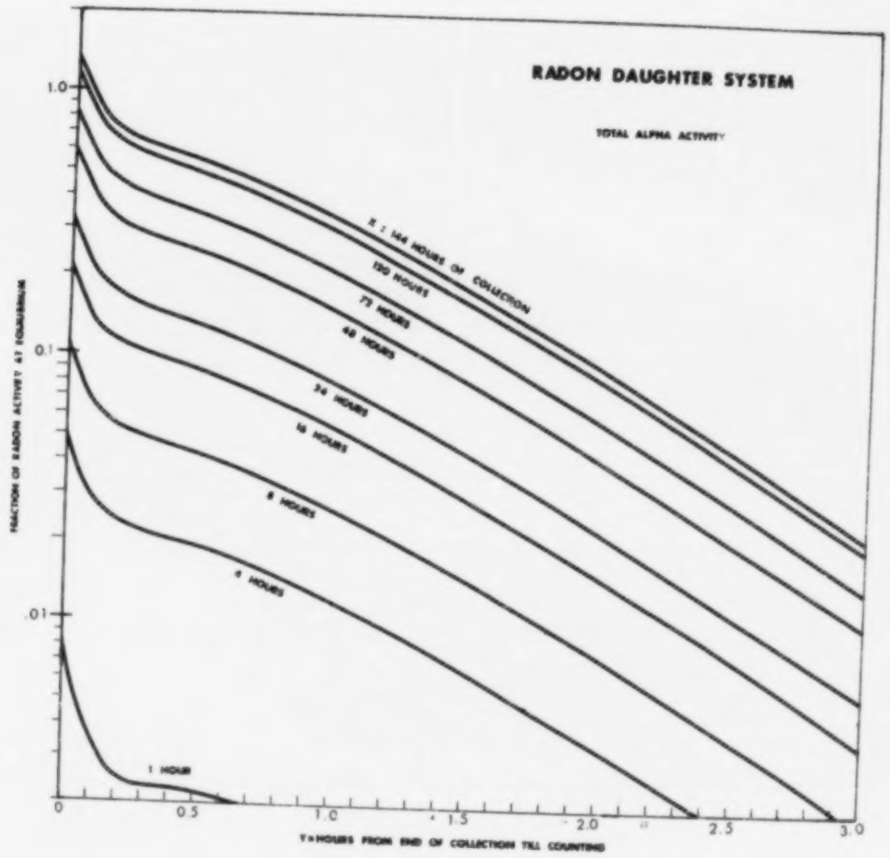

Figure 2. This is a plot of extrapolation factor E_{IIA} as a function of the elapsed time y. The plot presents typical ingrowth or leak testing times x from 1 to 144 hours. This graph is applicable when a radium leak test method is used that collects only the radon daughter products and when the sample is counted for alpha activity. For example, this is applicable to the method described by Benson and Fetz (Am. J. Roentgenol., Radium Therapy Nucl. Med., Vol. XCIX, No. 2, February 1967). The extrapolation factor decreases more rapidly than in Figure 1, because only the radon daughter products are present in the sample, and these have a short half life. (Radium B has a half life of 26.8 minutes.) Unlike the situation in Figure 1, it is therefore important that the sample be counted as soon after collection as possible. The plot is a graphic presentation of the data contained in Table II-A.

Table I-A. This is a table of the extrapolation factor E_{IA} for the total alpha activity of the radon plus daughter system of radium leak testing.

Y = Hours From End of Collection Till Counting	X = HOURS OF COLLECTION							
	1	4	8	16	24	48	72	144
0.0	.0160	.0799	.1667	.3328	.4891	.9052	1.2524	1.9852
1.0	.0202	.0858	.1720	.3368	.4920	.9050	1.2495	1.9768
2.0	.0217	.0874	.1729	.3365	.4905	.9004	1.2423	1.9642
3.0	.0219	.0873	.1722	.3346	.4874	.8942	1.2336	1.9500
4.0	.0219	.0868	.1711	.3322	.4839	.8876	1.2244	1.9354
5.0	.0217	.0862	.1698	.3297	.4802	.8810	1.2153	1.9209
6.0	.0216	.0855	.1685	.3272	.4766	.8743	1.2061	1.9065
7.0	.0214	.0849	.1673	.3248	.4731	.8678	1.1970	1.8921
8.0	.0213	.0842	.1660	.3223	.4695	.8612	1.1880	1.8779
9.0	.0211	.0836	.1648	.3199	.4660	.8548	1.1791	1.8638
10.0	.0209	.0830	.1635	.3175	.4625	.8483	1.1702	1.8498
11.0	.0208	.0823	.1623	.3151	.4590	.8419	1.1614	1.8359
12.0	.0206	.0817	.1611	.3127	.4555	.8356	1.1527	1.8220
13.0	.0205	.0811	.1599	.3104	.4521	.8293	1.1440	1.8083
14.0	.0203	.0805	.1587	.3081	.4487	.8231	1.1354	1.7947
15.0	.0202	.0799	.1575	.3057	.4453	.8169	1.1269	1.7812
16.0	.0200	.0793	.1563	.3034	.4420	.8108	1.1184	1.7678
17.0	.0199	.0787	.1551	.3012	.4387	.8047	1.1100	1.7545
18.0	.0197	.0781	.1539	.2989	.4354	.7986	1.1016	1.7413
19.0	.0196	.0775	.1528	.2966	.4321	.7926	1.0934	1.7282
20.0	.0194	.0769	.1516	.2944	.4288	.7866	1.0851	1.7152
21.0	.0193	.0764	.1505	.2922	.4256	.7807	1.0770	1.7023
22.0	.0191	.0758	.1494	.2900	.4224	.7748	1.0689	1.6895
23.0	.0190	.0752	.1482	.2878	.4192	.7690	1.0608	1.6768
24.0	.0188	.0746	.1471	.2857	.4161	.7632	1.0529	1.6642
25.0	.0187	.0741	.1460	.2835	.4129	.7575	1.0449	1.6517
26.0	.0186	.0735	.1449	.2814	.4098	.7518	1.0371	1.6393
27.0	.0184	.0730	.1438	.2793	.4068	.7461	1.0293	1.6269
28.0	.0183	.0724	.1427	.2772	.4037	.7405	1.0215	1.6147
29.0	.0181	.0719	.1417	.2751	.4007	.7350	1.0138	1.6026
30.0	.0180	.0713	.1406	.2730	.3976	.7294	1.0062	1.5905
31.0	.0179	.0708	.1395	.2709	.3947	.7239	.9986	1.5785
32.0	.0177	.0703	.1385	.2689	.3917	.7185	.9911	1.5667
33.0	.0176	.0697	.1374	.2669	.3887	.7131	.9837	1.5549
34.0	.0175	.0692	.1364	.2649	.3858	.7077	.9763	1.5432
35.0	.0173	.0687	.1354	.2629	.3829	.7024	.9689	1.5316
36.0	.0172	.0682	.1344	.2609	.3800	.6971	.9617	1.5201
37.0	.0171	.0677	.1334	.2589	.3772	.6919	.9544	1.5086
38.0	.0169	.0672	.1324	.2570	.3743	.6867	.9472	1.4973
39.0	.0168	.0666	.1314	.2551	.3715	.6815	.9401	1.4860
40.0	.0167	.0661	.1304	.2531	.3687	.6764	.9330	1.4748
41.0	.0166	.0656	.1294	.2512	.3659	.6713	.9260	1.4637
42.0	.0164	.0652	.1284	.2493	.3632	.6662	.9191	1.4527
43.0	.0163	.0647	.1274	.2475	.3605	.6612	.9121	1.4418
44.0	.0162	.0642	.1265	.2456	.3578	.6563	.9053	1.4310
45.0	.0161	.0637	.1255	.2438	.3551	.6513	.8985	1.4202
46.0	.0159	.0632	.1246	.2419	.3524	.6464	.8917	1.4095
47.0	.0158	.0627	.1237	.2401	.3497	.6416	.8850	1.3989
48.0	.0157	.0623	.1227	.2383	.3471	.6367	.8784	1.3884

Table I-B. This is a table of the extrapolation factor E_{IB} for the total beta activity of the radon plus daughter system of radium leak testing.

Y = Hours From End of Collection Till Counting	X = HOURS OF COLLECTION							
	1	4	8	16	24	48	72	144
0.0	.0048	.0454	.1035	.2147	.3193	.5978	.8302	1.3207
1.0	.0118	.0553	.1130	.2233	.3272	.6036	.8342	1.3210
2.0	.0141	.0580	.1152	.2247	.3278	.6022	.8311	1.3142
3.0	.0146	.0583	.1152	.2238	.3261	.5984	.8256	1.3051
4.0	.0146	.0580	.1145	.2223	.3238	.5941	.8195	1.2955
5.0	.0145	.0576	.1136	.2207	.3214	.5897	.8134	1.2858
6.0	.0144	.0572	.1128	.2190	.3190	.5852	.8073	1.2761
7.0	.0143	.0568	.1119	.2174	.3166	.5808	.8012	1.2665
8.0	.0142	.0564	.1111	.2157	.3142	.5765	.7952	1.2570
9.0	.0141	.0559	.1103	.2141	.3119	.5721	.7892	1.2475
10.0	.0140	.0555	.1094	.2125	.3095	.5678	.7833	1.2381
11.0	.0139	.0551	.1086	.2109	.3072	.5635	.7774	1.2288
12.0	.0138	.0547	.1078	.2093	.3049	.5593	.7715	1.2196
13.0	.0137	.0543	.1070	.2077	.3026	.5551	.7657	1.2104
14.0	.0136	.0539	.1062	.2062	.3003	.5509	.7600	1.2013
15.0	.0135	.0535	.1054	.2046	.2981	.5468	.7543	1.1923
16.0	.0134	.0531	.1046	.2031	.2958	.5427	.7486	1.1833
17.0	.0133	.0527	.1038	.2016	.2936	.5386	.7430	1.1744
18.0	.0132	.0523	.1030	.2000	.2914	.5345	.7374	1.1656
19.0	.0131	.0519	.1022	.1985	.2892	.5305	.7318	1.1568
20.0	.0130	.0515	.1015	.1971	.2870	.5265	.7263	1.1481
21.0	.0129	.0511	.1007	.1956	.2849	.5226	.7209	1.1395
22.0	.0128	.0507	.1000	.1941	.2827	.5186	.7154	1.1309
23.0	.0127	.0503	.0992	.1926	.2806	.5147	.7101	1.1224
24.0	.0126	.0499	.0985	.1912	.2785	.5109	.7047	1.1139
25.0	.0125	.0496	.0977	.1897	.2764	.5070	.6994	1.1056
26.0	.0124	.0492	.0970	.1883	.2743	.5032	.6942	1.0972
27.0	.0123	.0488	.0962	.1869	.2722	.4994	.6889	1.0890
28.0	.0122	.0485	.0955	.1855	.2702	.4957	.6838	1.0808
29.0	.0121	.0481	.0948	.1841	.2682	.4919	.6786	1.0727
30.0	.0120	.0477	.0941	.1827	.2661	.4882	.6735	1.0646
31.0	.0119	.0474	.0934	.1813	.2641	.4846	.6684	1.0566
32.0	.0119	.0470	.0927	.1800	.2622	.4809	.6634	1.0486
33.0	.0118	.0467	.0920	.1786	.2602	.4773	.6584	1.0408
34.0	.0117	.0463	.0913	.1773	.2582	.4737	.6535	1.0329
35.0	.0116	.0460	.0906	.1759	.2563	.4701	.6485	1.0252
36.0	.0115	.0456	.0899	.1746	.2544	.4666	.6437	1.0174
37.0	.0114	.0453	.0892	.1733	.2524	.4631	.6388	1.0098
38.0	.0113	.0449	.0886	.1720	.2505	.4596	.6340	1.0022
39.0	.0112	.0446	.0879	.1707	.2487	.4562	.6293	.9947
40.0	.0112	.0443	.0872	.1694	.2468	.4527	.6245	.9872
41.0	.0111	.0439	.0866	.1682	.2449	.4493	.6198	.9798
42.0	.0110	.0436	.0859	.1669	.2431	.4459	.6152	.9724
43.0	.0109	.0433	.0853	.1656	.2413	.4426	.6105	.9651
44.0	.0108	.0429	.0846	.1644	.2394	.4393	.6059	.9578
45.0	.0107	.0426	.0840	.1631	.2376	.4359	.6014	.9506
46.0	.0107	.0423	.0834	.1619	.2359	.4327	.5969	.9435
47.0	.0106	.0420	.0828	.1607	.2341	.4294	.5924	.9364
48.0	.0105	.0417	.0821	.1595	.2323	.4262	.5879	.9293

Table II-A. This is a table of the extrapolation factor E_{IIA} for the total alpha activity of the radon daughter system of radium leak testing.

Y = Hours From End of Collection Till Counting	X = HOURS OF COLLECTION							
	1	4	8	16	24	48	72	144
0.0	.0084	.0502	.1081	.2190	.3234	.6013	.8331	1.3224
.1	.0036	.0290	.0653	.1348	.2002	.3743	.5195	.8261
.2	.0025	.0235	.0536	.1113	.1656	.3101	.4306	.6851
.3	.0023	.0216	.0491	.1018	.1514	.2835	.3937	.6263
.4	.0023	.0203	.0459	.0950	.1412	.2641	.3667	.5832
.5	.0022	.0190	.0428	.0883	.1311	.2452	.3403	.5411
.6	.0021	.0177	.0395	.0814	.1208	.2258	.3133	.4981
.7	.0020	.0162	.0362	.0744	.1104	.2062	.2862	.4549
.8	.0018	.0148	.0329	.0676	.1002	.1871	.2596	.4125
.9	.0017	.0134	.0297	.0610	.0904	.1687	.2340	.3719
1.0	.0015	.0121	.0267	.0547	.0811	.1513	.2099	.3336
1.1	.0014	.0108	.0239	.0489	.0725	.1352	.1875	.2979
1.2	.0013	.0097	.0213	.0435	.0645	.1202	.1668	.2650
1.3	.0011	.0086	.0189	.0386	.0572	.1066	.1479	.2349
1.4	.0010	.0076	.0167	.0341	.0506	.0942	.1307	.2076
1.5	.0009	.0067	.0147	.0301	.0446	.0831	.1152	.1830
1.6	.0008	.0059	.0130	.0265	.0392	.0731	.1013	.1610
1.7	.0007	.0052	.0114	.0233	.0344	.0641	.0889	.1413
1.8	.0006	.0045	.0100	.0204	.0302	.0562	.0779	.1238
1.9	.0005	.0040	.0087	.0178	.0264	.0491	.0681	.1082
2.0	.0004	.0035	.0076	.0156	.0230	.0429	.0595	.0945
2.1	.0004	.0030	.0066	.0136	.0201	.0374	.0519	.0824
2.2	.0003	.0026	.0058	.0118	.0175	.0326	.0452	.0718
2.3	.0003	.0023	.0050	.0103	.0152	.0283	.0393	.0624
2.4	.0002	.0020	.0044	.0089	.0132	.0246	.0341	.0542
2.5	.0002	.0017	.0038	.0077	.0114	.0214	.0296	.0471
2.6	.0002	.0015	.0033	.0067	.0099	.0185	.0257	.0408
2.7	.0001	.0013	.0028	.0058	.0086	.0160	.0222	.0353
2.8	.0001	.0011	.0024	.0050	.0074	.0139	.0193	.0306
2.9	.0001	.0009	.0021	.0043	.0064	.0120	.0166	.0265
3.0	.0001	.0008	.0018	.0037	.0055	.0104	.0144	.0229
3.1	.0001	.0007	.0016	.0032	.0048	.0090	.0124	.0198
3.2	.0000	.0006	.0013	.0028	.0041	.0077	.0107	.0171
3.3	.0000	.0005	.0012	.0024	.0036	.0067	.0093	.0147
3.4	.0000	.0004	.0010	.0021	.0031	.0057	.0080	.0127
3.5	.0000	.0004	.0008	.0018	.0026	.0049	.0069	.0109
3.6	.0000	.0003	.0007	.0015	.0023	.0043	.0059	.0094
3.7	.0000	.0003	.0006	.0013	.0019	.0037	.0051	.0081
3.8	.0000	.0002	.0005	.0011	.0017	.0031	.0044	.0070
3.9	.0000	.0002	.0004	.0010	.0014	.0027	.0038	.0060
4.0	.0000	.0001	.0004	.0008	.0012	.0023	.0032	.0052
4.1	.0000	.0001	.0003	.0007	.0010	.0020	.0028	.0044
4.2	.0000	.0001	.0003	.0006	.0009	.0017	.0024	.0038
4.3	.0000	.0001	.0002	.0005	.0008	.0015	.0020	.0033
4.4	.0000	.0001	.0002	.0004	.0006	.0012	.0017	.0028
4.5	.0000	.0000	.0001	.0004	.0005	.0011	.0015	.0024
4.6	.0000	.0000	.0001	.0003	.0005	.0009	.0013	.0021
4.7	.0000	.0000	.0001	.0002	.0004	.0008	.0011	.0018
4.8	.0000	.0000	.0001	.0002	.0003	.0007	.0009	.0015

Table II-B. This is a table of the extrapolation factor E_{IIB} for the total beta[12] activity of the radon daughter system of radium leak testing.

Y = Hours From End of Collection Till Counting	X = HOURS OF COLLECTION							
	1	4	8	16	24	48	72	144
0.0	.0048	.0454	.1035	.2147	.3193	.5978	.8302	1.3207
.1	.0052	.0447	.1007	.2079	.3087	.5771	.8011	1.2738
.2	.0051	.0420	.0940	.1935	.2871	.5364	.7443	1.1833
.3	.0048	.0387	.0863	.1772	.2628	.4908	.6809	1.0822
.4	.0045	.0353	.0784	.1608	.2384	.4450	.6173	.9811
.5	.0041	.0320	.0708	.1450	.2149	.4009	.5561	.8837
.6	.0038	.0288	.0635	.1301	.1927	.3594	.4984	.7919
.7	.0034	.0258	.0568	.1161	.1720	.3207	.4447	.7066
.8	.0031	.0230	.0505	.1033	.1529	.2851	.3954	.6281
.9	.0027	.0204	.0448	.0916	.1355	.2526	.3503	.5565
1.0	.0024	.0181	.0396	.0809	.1198	.2232	.3095	.4917
1.1	.0022	.0159	.0350	.0713	.1056	.1967	.2727	.4332
1.2	.0019	.0140	.0308	.0627	.0928	.1729	.2398	.3809
1.3	.0017	.0123	.0270	.0550	.0815	.1517	.2104	.3342
1.4	.0015	.0108	.0237	.0482	.0713	.1329	.1842	.2926
1.5	.0013	.0095	.0207	.0422	.0624	.1162	.1611	.2558
1.6	.0011	.0083	.0181	.0368	.0544	.1014	.1406	.2233
1.7	.0010	.0072	.0157	.0321	.0475	.0884	.1226	.1947
1.8	.0008	.0063	.0137	.0279	.0413	.0770	.1067	.1695
1.9	.0007	.0055	.0119	.0243	.0359	.0669	.0928	.1474
2.0	.0006	.0047	.0104	.0211	.0312	.0581	.0806	.1280
2.1	.0005	.0041	.0090	.0183	.0271	.0504	.0699	.1111
2.2	.0005	.0036	.0078	.0159	.0235	.0437	.0606	.0963
2.3	.0004	.0031	.0067	.0137	.0203	.0379	.0525	.0834
2.4	.0003	.0027	.0058	.0119	.0176	.0328	.0455	.0722
2.5	.0003	.0023	.0050	.0103	.0152	.0284	.0393	.0625
2.6	.0002	.0020	.0043	.0089	.0132	.0245	.0340	.0540
2.7	.0002	.0017	.0038	.0077	.0114	.0212	.0294	.0467
2.8	.0002	.0015	.0032	.0066	.0098	.0183	.0254	.0403
2.9	.0001	.0013	.0028	.0057	.0085	.0158	.0219	.0348
3.0	.0001	.0011	.0024	.0049	.0073	.0136	.0189	.0300
3.1	.0001	.0009	.0021	.0042	.0063	.0117	.0163	.0258
3.2	.0001	.0008	.0018	.0036	.0054	.0101	.0140	.0223
3.3	.0001	.0007	.0015	.0031	.0046	.0087	.0121	.0192
3.4	.0000	.0006	.0013	.0027	.0040	.0075	.0104	.0165
3.5	.0000	.0005	.0011	.0023	.0034	.0064	.0089	.0142
3.6	.0000	.0004	.0010	.0020	.0029	.0055	.0077	.0122
3.7	.0000	.0003	.0008	.0017	.0025	.0047	.0066	.0105
3.8	.0000	.0003	.0007	.0015	.0022	.0041	.0057	.0090
3.9	.0000	.0002	.0006	.0012	.0019	.0035	.0049	.0078
4.0	.0000	.0002	.0005	.0011	.0016	.0030	.0042	.0067
4.1	.0000	.0002	.0004	.0009	.0014	.0026	.0036	.0057
4.2	.0000	.0001	.0004	.0008	.0012	.0022	.0031	.0049
4.3	.0000	.0001	.0003	.0007	.0010	.0019	.0026	.0042
4.4	.0000	.0001	.0002	.0006	.0008	.0016	.0023	.0036
4.5	.0000	.0001	.0002	.0005	.0007	.0014	.0019	.0031
4.6	.0000	.0001	.0002	.0004	.0006	.0012	.0016	.0026
4.7	.0000	.0000	.0001	.0003	.0005	.0010	.0014	.0023
4.8	.0000	.0000	.0001	.0003	.0004	.0009	.0012	.0019

Table III. The values of half-life shown in the table of the radium decay series were used to calculate the extrapolation factors found in the previous tables.

SYMBOL	HISTORICAL NAME	ENERGY AND PERFECT ABUNDANCE OF RADIATION EMITTED			HALF LIFE
		Alpha	Beta	Gamma	
^{226}Ra	Radium	4.79 (98.8%) 4.61 (1.2%)		0.18 (1.2%)	1622 y.
^{222}Rn	Radon	5.49			3.83 d.
^{218}Po	Radium-A	6.00			3.05 min.
^{214}Pb	Radium-B		0.65	.241 (11.5%) .294 (25.8%) .350 (45.0%)	26.8 min.
^{214}Bi	Radium-C		3.17 (23%) 1.7 (77%)	.607 (65.8%) .766 (6.5%) .933 (6.7%) 1.120 (20.6%) 1.238 (6.3%) 1.379 (6.4%) 1.761 (25.8%) 2.198 (7.4%)	19.7 min.
^{214}Po	Radium-C'	7.68			164 μ sec.

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